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Emission properties study of a Nd3+ doped TZA glass random laser

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ABSTRACT

Random lasers (RLs) have been thoroughly studied for applications such as high definition speckle free imaging, lithography, miniature spectroscopy, etc. RLs made with crystalline powders have shown promising results, with high emission efficiencies and narrow wavelength bandwidth. However, few studies on glass random lasers have been made, since its inhomogeneous broadening make it hard to verify the linewidth narrowing characteristic of laser emission. Here, we describe linewidth and temporal measurements for a TZA glass doped with 16 wt% neodymium. We verified a 0.5 nm linewidth narrowing at laser threshold. The pump intensity where the transition occurs coincided with the appearance of a faster emission decay, showing the presence of laser emission for higher pump power energies. This result is promising in understanding random lasing for glass powders.

Keywords: Random laser, glasses, temporal studies, rare-earth doped glass.

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1. INTRODUCTION

Random lasers (RLs) were proposed theoretically for the first time in the 1960s by Letokhov[1], having its first experimental demonstration in 1993 by Gouedard et al.[2] In these lasers, the stimulated emission is caused by scattering between micro/nanoparticles, or due to differences in the refractive index within the sample[3], differently from regular lasers where the engineering of the cavity is responsible for the emission gain. Therefore, random lasers are cheaper and easier to fabricate than regular lasers, and have potential applications such as remote sensing, encrypting, cancer detection, among others.

RLs have been demonstrated with dyes[4], fibers[5], crystal powders[6], polymers[7] and nanoparticles doped with rare-earth ions [8]. Particularly, high efficiency random lasers have been demonstrated with Neodymium (Nd) doped materials [6], which shows the promising capabilities of this kind of rare-earth random laser. The standard proof for random laser action is obtained through the observation of its linewidth narrowing when it reaches the stimulated emission regime, as well as the slope increase of its power emission curve after threshold.

Glasses are materials that can be easily doped with rare-earths and have different characteristics depending on their matrix. They can have high refractive index, large mechanical resistance, and are easy and usually cheap to fabricate. Glass random lasers, however, have rarely been studied [9, 10], likely due to its inhomogeneous emission band [11] and easily damageable surface caused by high pump power. The inhomogeneous emission makes the linewidth narrowing difficult to observe, which is the main method used to verify random lasing, and the low surface damage threshold can make the material unsuitable to reach the pump powers beyond laser threshold. One method to verify random lasing in these kinds of materials involves the study of their temporal behavior. By measuring the emission decay time, it is possible to obtain the spontaneous decay time, and if a second, shorter decay time appears, spontaneous emission will also be occurring, which demonstrates laser behavior. This kind of study has been done previously [12] for a crystal powder random laser doped with Nd^{3+}, showing good results.

Here, we present a spectral and a temporal study of a tellurite glass with aluminum oxide doped with 16 wt% Nd powder pellet, with two different powder sizes (20 to 45 μm and 45 to 75 μm).

It was not possible to obtain conclusive results by observing only the linewidth narrowing due to a spectrometer imprecision. Nevertheless, a 0.5 nm spectral narrowing for larger pump energies per area than 0.5 mJ/mm^2 was observed

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in both studied samples. To corroborate this result, a time decay measurement was made, showing the appearance of a shorter decay time for the samples around 0.59 mJ/mm^2, demonstrating laser action with approximately 30% of stimulated emission.

2. MATERIALS AND METHODS

The glass was prepared at the Faculdade de Tecnologia de Sao Paulo, using a melt-quenching method adding 16 wt% of Nd_2O_3 to the sample composition: 85.0 TeO_2-12.95 ZnO-2.05 Al_2O_3 (TZA). The characterization of this sample was previously published for the case of 0.5 wt% neodymium doping[13], showing good absorption at 806 nm and a strong emission at 1064 nm.

The TZA was turned into powder by using an agate pestle, and the particles were separated into two different size ranges using alumina sieves: from 20 to 45 μm and from 45 to 75 μm. Since there was no washing process, the resulting powders were polydispersed in size, which was purposefully chosen due to previous published results [6] that demonstrated high efficient random lasers of a neodymium doped material in this condition. These powders were pressed into 7 mm diameter and 1 mm height pellets using 255 MPa of pressure.

Two different measurements were made with these pellets: linewidth narrowing and temporal emission. For both kinds of measurements, an OPOTek model OPOlette laser system was used, with 10 ns pulses at a repetition rate of 20 Hz, emitting at 806 nm. The experimental setup used is shown in Figure 1. For temporal measurements, an oscilloscope was used to verify the decay time of the studied sample, while for the linewidth measurement, an OceanOptics® spectrometer connected to a computer with the SpectraSuite® program installed was used to obtain the emission spectra.

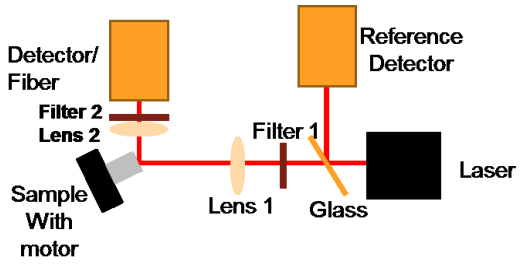

Figure 1. Experimental setup. It consists of the pump laser, which is reflected by a beam splitter (~4% reflectivity) into a reference detector for time measurements; a longpass filter of 700 nm to let only the 806 nm pump wavelength pass; a 15 cm focal length lens (Lens 1), which focalizes the beam over the sample located on a rotating motor; a 2.5 cm focal length lens (Lens 2), which focalizes the emitted beam into either a photodetector or a fiber for temporal and linewidth measurements respectively; and a longpass filter (Filter 2), which blocks wavelengths lower than 1000 nm from reaching the detector.

The spectra were measured for each pump intensity, with integration times varying from 200 ms to 500 ms, with 50 averages per measurement to reduce noise due to laser fluctuations. A Gaussian fit was done for each spectrum, centered in ~1064 nm, to obtain the full width half maximum (FWHM) and verify its changes for different pump energies.

For the temporal studies, we considered that the time decays were an exponential sum of the fast and the slow decay times, making fits with the following equation:

$$\text{Emission} = Ae^{\frac{-x}{\tau_1}} + Be^{\frac{-x}{\tau_2}} \quad (1)$$

, where A and τ_1 are related to the stimulated emission contribution, which only appears when there is laser emission, and B and τ_2 are related to the spontaneous emission. From this fitting, it was possible to calculate the fraction of stimulated emission as a function of pump power [12], A/(A+B), which will quantify the percentage of stimulated emission in relation to the total measured power.

3. RESULTS

3.1 Linewidth measurements

Linewidth measurements were made using an OceanOptics® Spectrometer with 0.6 nm resolution. The incident energy was varied from 0.1 mJ up to 1.4 mJ in a circular focus area of 0.83 mm^2. Results are shown in Figure 2.

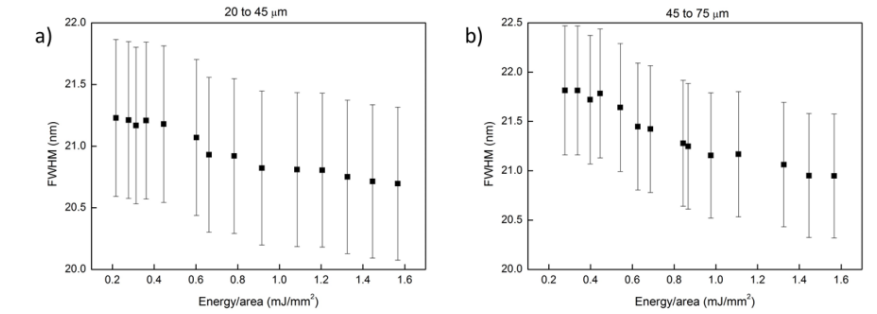

Figure 2: Linewidth measurements for pellets with a) 20 to 45 μm; and b) 45 to 75 μm powder sizes.

A 0.5 nm difference between the largest (~21.9 nm for 20 to 45 μm and ~22.5 nm for 45 to 75 μm) and smallest (~21.4 nm for 20 to 45 μm and 21.6 nm for 45 to 75 μm) measured FWHMs is observed in Figure 2. However, this difference is smaller than the error bars for the measurements, and, therefore, is not conclusive in this case.

After further investigating the linewidth narrowing with another, higher precision spectrometer, we verified that this result was not reproducible. By investigating the origin of this behavior in the first used spectrometer, we found that an anomalous behavior occurred only in the 1064 nm wavelength region, where the shape of the spectrum changed as a function of power, with the FWHM increasing from 0.4 nm to 1.2 nm in this specific wavelength region. The equipment was sent to the manufacturer for inspection, where this behavior showed to be reproducible but no explanation for this was found. Therefore, we discarded this measurement and proceeded with the temporal studies to find proof of laser action for the glass samples.

3.2 Temporal measurements

Time decay measurements were taken from pump energies between 0.16 mJ and 1.5 mJ with the same 0.83 mm^2 circular pump area. One typical result is shown in Figure 3a for the 20 to 45 μm pellet, with three different pump energies. It is clear that the curve becomes steeper for larger pump energies, which is expected if there is more than spontaneous emission present in the signal.

Double exponential fittings were made in these curves using Equation (1). For low pump energies, only single exponential fittings were obtained, with a single decay time value. For higher energies, two different exponentials were fitted, with different times as well as A and B factors both positive. In Figure 3b, we compare a single exponential fitting (blue) with a double exponential fitting (red). In the first microseconds of emission, the single exponential fitting remains outside of the emission curve, clearly showing the presence of a second decay time for this pump energy.

We fitted a single exponential line to obtain the spontaneous emission decay time of 6.1 μs in both samples for the lowest pump energy of 0.1 mJ. As the energy increased, this long decay time remained approximately the same, and for energies per area larger than 0.59 mJ/mm^2, a second exponential decay appeared for 20 to 45 μm as well as for 45 to 75 μm, indicating stimulated emission. Comparing this threshold with the one observed in the linewidth narrowing one (0.5 mJ/mm^2), there was an 18% difference between them, which is acceptable considering laser energy fluctuations during both kinds of measurements. The short decay time varied from 3.73 μs to 2.23 μs for the smaller powder size pellet, and from 4.11 μs to 2.15 μs for the larger one, getting smaller with higher pump energy as shown in Figure 4. The smaller the second decay time, the larger the factor A, which is related to an increase in the stimulated emission.

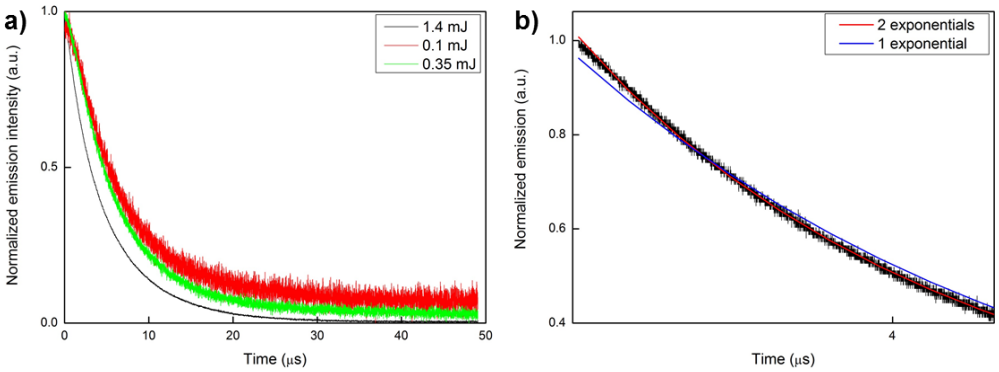

Figure 3: a) Normalized time decay measurements for three different pump energies for the 20 to 45 μm pellet; b) Comparison between a single exponential (blue) and a double exponential (red) fitting for the time decay with 1.4 mJ pump energy for the 20 to 45 μm pellet.

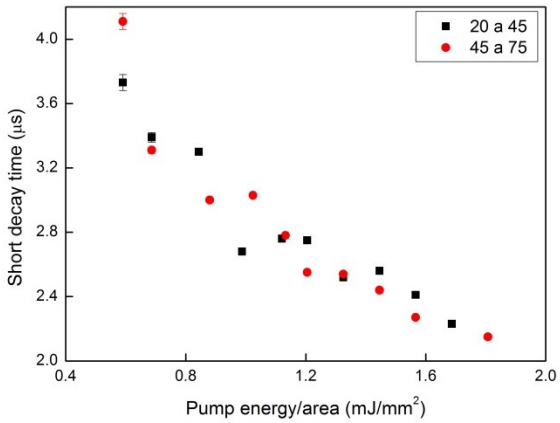

Figure 4: Short decay time for different pump energies for the 20 to 45 μm (black squares) and 45 to 75 μm (red balls) pellets.

From the double exponential fittings, we obtained the A and B factors for each powder size pellet and calculated the ratio between stimulated and spontaneous emission, given by A/(A+B). In Figure 5, the variation of A/(A+B) can be seen for both studied powder sizes. For the 20 to 45 μm powder, the stimulated emission started around 23% and increased to 39% for the maximum energy per area (1.7 mJ/mm^2), while the 45 to 75 μm one had approximately 22% of stimulated emission and increased to 33% at 1.8 mJ/mm^2. This shows that both pellets present laser behavior, with the 20 to 45 μm one giving a higher stimulated emission ratio than the 45 to 75 μm one.

Since A/(A+B) shows an increasing behavior up to the maximum energy available, it is likely that if the pump energy is further increased, even more stimulated emission would be seen, and better efficiency would be reached. This was not studied since for pump energies/area higher than 1.2 mJ/mm^2 there was damage to the surface of the pellets, making it a limitation for this random laser material.

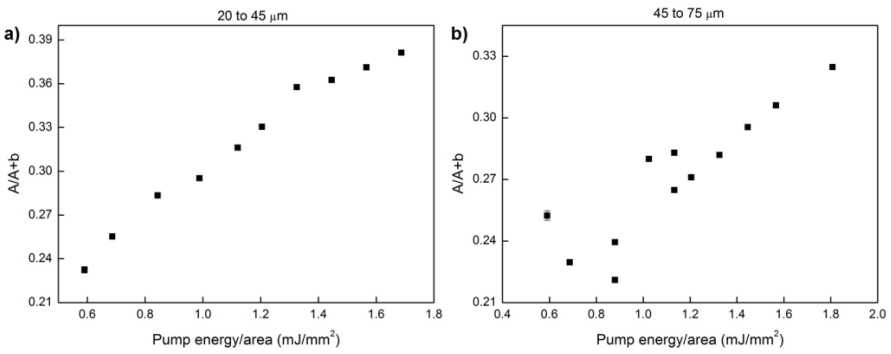

Figure 5: Stimulated emission ratio for a) 20 to 45 μm and b) 45 to 75 μm pellet.

4. CONCLUSION

Two different sizes of powder pellets of a TZA glass doped with 16 wt% Nd were studied. Linewidth measurements showed a 0.5 nm narrowing for pump energies/area larger than 0.5 mJ/mm^2, though due to the equipment imprecision this result is not reliable and was discarded. Temporal measurements were made to verify this, and they show the presence of a second, smaller time decay for pump energies/area larger than 0.59 mJ/mm^2, proving that laser emission occurs in these samples.

With both methods, it is possible to say that there is stimulated emission, though its efficiency is low (~30%) compared to other studied materials (~50% or higher[12]). This demonstrated laser activity for the first time for a TZA glass, and also provided a method that can be used to characterize random lasing behavior for other glasses by using only time measurements.

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