One- and two-photon pumped random laser action in Rhodamine B doped di-ureasil hybrid

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ABSTRACT

One- and two-photon pumped random lasing have been demonstrated in a powder based on a Rhodamine B-doped di-ureasil hybrid by using a picosecond pump laser emitting either at 532 nm or 1064 nm under controlled experimental conditions. In both cases, we have used the same diffusive medium with identical spatial disorder, the same time-width and temporal profile for the pump pulse, as well as equal pump spot sizes. The corresponding onsets of laser-like emission and slope efficiencies are also investigated.

Keywords: Laser materials, time-resolved spectroscopy, random lasing, ultrafast lasers

1. INTRODUCTION

Since first proposed by Letokhov in 1967 [1], lasing in random media has become a subject of intense theoretical and experimental studies due to the important scientific and technological implications of this new research field [2]. Random lasing has been observed in a wide range of scattering systems such as solutions of microparticles dispersed in a laser dye, neodymium doped crystal powders, ceramic and polymeric systems, semiconductor nanoparticles, organic tissues, liquid crystals, etc (see Refs. [2-4] and references therein). The nature and morphology of each amplifying disordered medium determine its specific feedback mechanism and random laser behavior making it difficult to study and compare all the previously mentioned systems by means of a unique theoretical treatment [5-11]. Only recently, the real-time behavior of solid state random laser systems both in the spectral and temporal domains has been explored [12-15].

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In this work, one (OP)- and two (TP)-photon pumped random lasing are studied in a Rhodamine B-doped di-ureasil hybrid by using a picosecond pump laser, emitting either at 532 nm or 1064 nm, under controlled experimental conditions. In these experiments, we use one diffusive medium with identical spatial disorder, the same time-width and temporal profile for the pump pulse, as well as equal pump spot sizes. Only under these conditions, reliable comparisons of the spectral and temporal dynamics of the random laser excited by one or two photons are possible.

Note that due to the covalent linkage between its organic and inorganic parts, a higher efficiency of the dye emission is expected if compared to hybrid materials based on silica gels where the dye molecules are only entangled in the porous silica network.

2. EXPERIMENTAL

2.1 Synthesis and characterization of the laser sample

The reagents O,O′-Bis(2-aminopropyl) polypropylene glycol-block-polyethylene glycol-block-polypropylene glycol (Fluka), commercially known as Jeffamine-ED600®, average molecular weight 600 g.mol^{-1}, 3-isocyanatepropyltriethoxysilane (ICPTES) (Aldrich 95%), ethyl alcohol absolute P.A. (Carlo Erba), tetrahydrofuran P.A. (stabilized - Riedel-de Haën), HCl (ACS Reagent 37% - Sigma-Aldrich) and Rhodamine B were used as received. The di-ureasil host, termed as d-U(600), contains 8.5 (OCH_2CH_2) polymer chains with both ends grafted to a siliceous network by means of urea linkages. The cross-links between the organic and the inorganic components were formed by reacting the NH_2 groups of Jeffamine-ED600® with the –N=C=O group of ICPTES, in THF, under magnetic stirring and reflux at 80 ^oC for 18 h. The non-hydrolyzed d-U(600) precursor was isolated after complete THF evaporation at 45 ^oC in a rotary bench evaporator. A solution of RhB chloride in 1 mL of ethanol was incorporated into the di-ureasil host, under magnetic stirring. The RhB solutions were added to 3 g of d-UPTES after 15 min. The suspensions were kept under magnetic stirring for 15 min at room temperature, and were then cast into a polystyrene mould (1×1×3 cm) and left to gel, which happened within 3 min. After gelation, the mould was covered by Parafilm® and kept at room temperature for 24 h. Then the cover was removed and a three-step heat treatment at 40 ^oC (72 h), 50 ^oC (24 h) and 60 ^oC (24 h) was performed to eliminate residual solvents (including ethanol and water produced by polycondensation). The final volume of the sample denoted by d-U-RhB15m was not significantly affected by shrinkage process (less than 5%). Afterwards, it was ground by using a mixer mill (Retsch MM200) during 4 min. The polydispersity of the measured powder was evaluated from Scanning Electron Microscopy finding an average particle size of 22 μm. The resulting powder was compacted in a 6 mm high cylindrical quartz cell with no front window for handling ease and optical characterization. The volume filling factor of the powder material (f = 0.55) was calculated by measuring sample volume and weight.

2.2 Experimental techniques

The random laser behavior found in d-U-RhB15m sample was studied by using the fundamental (1064 nm) and frequency doubled output (532 nm) of a 20 Hz, picoseconds mode-locked Nd:YAG laser as the excitation source. The laser excitation energy was controlled with a halfwave plate combined with a polarizer and measured with an energy meter (OPHIR: PE25 and PE10). The laser beam was focused in the sample to a spot size of 2.69 mm. The emitted radiation from the front face of the sample was collected with an optical fiber by use of two lenses. A short-pass filter and a long-pass filter were used to remove light at the pump frequencies. The one photon (OP) and two photons (TP) pumped random laser threshold of the studied sample was explored by spectrally resolving the emission signal with a fiber-coupled spectrometer (CVI SM240) and temporally analyzing it by a fast photodiode (Newport 1480-S) connected to a digital oscilloscope (Tektronix DP071604) as a function of the excitation energy.

3. RESULTS AND DISCUSSION

The random lasing properties of d-U-RhB15m powder such as pump power dependence of the emission spectra, emission kinetics, and laser-like emission threshold were studied after picosecond optical pumping (pump pulse duration of 30 ps) at 532 and 1064 nm.

Figure 1, grey dots, shows the effective emission linewidth ($\Delta\lambda_{eff} = \int \frac{I(\lambda)d\lambda}{I_{\max}}$) collapse obtained in this sample pumped at 532 nm upon increasing the pump pulse energy. From these experimental data, a laser threshold (defined as the energy value above which a suddenly drop of the spectral linewidth is observed) around 22 μJ/pulse was found. The black squares in Fig. 1 show the spectral narrowing of the emission obtained under two photon pumping at 1064 nm. As can be observed, in this case the threshold is around 4 mJ/pulse.

The temporal characteristics of the emitted pulse were also investigated. Figure 2 shows the output pulse shortening obtained as a function of the pump energy in the two photon pumping case. It is worthy mentioning that the full width at half maximum (FWHM) of the time profile was reduced down to around 100 ps above the onset of random laser emission. This value could be even much lower because the actual time resolution of the detection system, fast photodiode and broad band oscilloscope, used to perform this set of experiments is around 80 ps.

As can be seen, by comparing Figures 1 and 2, there is a fairly good agreement between the pump energy dependence of the spectral and temporal narrowings obtained at TP pumping.

In order to more precisely define the onset of the random lasing and the slope efficiency of the studied sample we have also performed the integration of the emitted intensity above threshold for the one and two photon pumped emissions. Figures 3 and 4 show the corresponding integrated intensities as a function of pump energy.

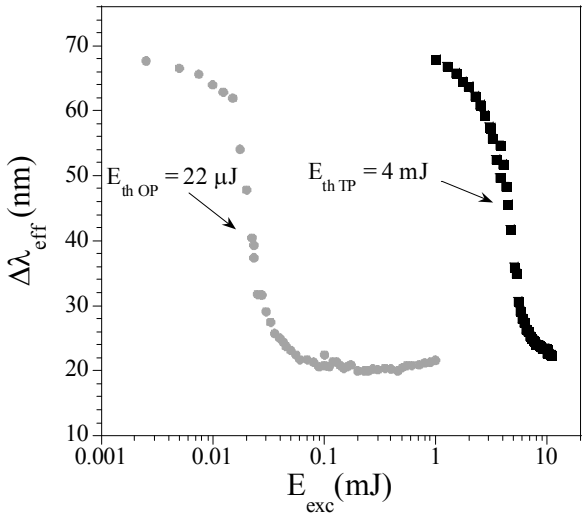

Fig. 1. Spectral narrowing of the emission obtained as a function of the pump energy for one (grey dots) and two (black squares) photon pumped random laser.

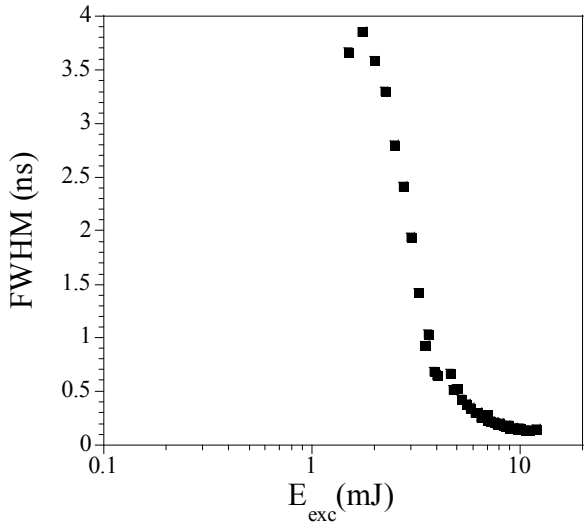

Fig. 2 Output pulse shortening obtained as a function of the pump energy in the TP case.

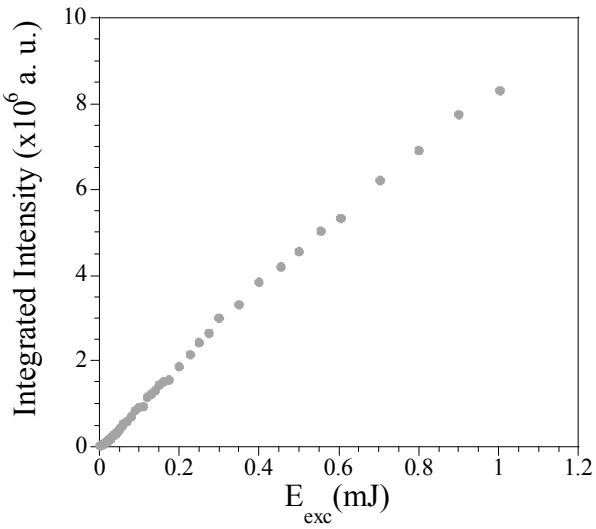

Fig. 3 Integrated intensity as a function of the pump energy with one photon pumping.

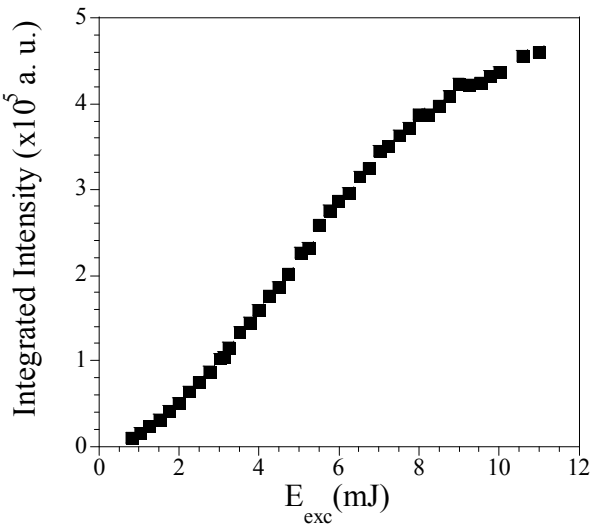

Fig. 4 Integrated intensity as a function of the pump energy for two photon pumping.

4. CONCLUSIONS

We have experimentally demonstrated the possibility of obtaining random lasing by pumping at one and two photons in a sample based in Rhodamine B doped di-ureasil with static disorder. The threshold for one and two photon pumping are 22 μJ and 4 mJ respectively. Although the threshold under one photon pumping seems to be similar to the one obtained in silica gel matrices, the results under two photon pumping deserve a more careful investigation to determine the influence of the two photon absorption cross section of Rhodamine B in both matrices in random lasing processes. More extensive studies are underway to clarify this point.

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