Random laser generation in rhodamine-doped aerogel

Adriana Ramos de Miranda*, Édison Pecoraro, Sidney José Lima Ribeiro**, Júlia Maria Giehl*, Saara-Maarit Reijn*, Niklaus Ursus Wetter***

** Centro de Lasers e Aplicações, CNEN-IPEN/SP, Av. Prof. Lineu Prestes, 2242*
Cidade Universitária - CEP 05508-000 São Paulo – SP - Brasil
*** Universidade Estadual Paulista Júlio de Mesquita Filho - Instituto de Química de Araraquara*
Rua Prof. Francisco Degni, 55 - Jardim Quitandinha – CEP 14800-900 - Araraquara – SP - Brasil
a.r.miranda@terra.com.br

Abstract: Random Laser generation can give valuable information on the structure of diffuse materials. We demonstrate for the first time random laser generation from an aerogel matrix doped with rhodamine 6G.

OCIS codes: (140.0140) Lasers and laser optics; (140.3380) Laser materials.

1. Introduction

The study of the optical signal of random lasers (RL) can provide accurate information about the structure of the scattering material [1, 2]. Therefore, disordered materials that can be produced by controlling the characteristics of the micro-structure itself are of great interest for the development and comprehension of the phenomenon [3]. In this context the laser generation from aerogel matrix doped with rhodamine 6G could be a promising methodology.

Silica aerogel is a material well known by its good mechanical and thermal properties. On the other hand, its optical properties are poor, since the extinction coefficient in the visible wavelength range is dominated by Rayleigh scattering [4]. Because such scattering is not coherent, low-density silica aerogel (LDSA) can be used as host for random lasers systems. To the best of our knowledge, it is the first time that monoliths of LDSA are used as scattering matrix for rhodamine 6G based random laser.

2. Methods

2.1. Rhodamine 6G Silica Aerogels synthesis

The reagents tetraethyl orthosilicate (TEOS) (Aldrich 98%), ethyl alcohol (ACS reagent, ≥99.5%, absolute - Sigma-Aldrich), hydrochloric acid (ACS Reagent 37% - Sigma-Aldrich) ammonium hydroxide solution (ACS reagent, 28.0-30.0% NH_3 basis - Sigma-Aldrich) and Rhodamine 6G hydrochloride (95% - Sigma) were used. The two-step catalyzed TEOS based doped xerogels were prepared from TEOS, ethanol, HCl and NH_4OH. In the first step, TEOS, deionized water, ethyl alcohol, HCl and Rhodamine 6G were mixed under magnetic stirring (molar ratio of $1:9:4:8.10^{-3}:2.5.10^{-4}$), for hydrolysis at room temperature during 60min. Next, NH_4OH (molar ratio $1:16.10^{-3}$) was added to the sol, shifting the pH from 2 to 8 for a rapid polycondensation reaction (about 40s). Before the gelation occurred, the sol was transferred to polystyrene molds (1x1x4cm), which were kept covered at room temperature for 14 days for aging. After that, the monoliths were transferred into a homemade CO_2 critical point drier, where the solvents (water and ethanol) were removed by cycling the samples above the supercritical conditions (35ºC and 1300psi) by 3 periods of 60min. The exhaustion rate of CO_2 was 8-10 SCFH. To remove any residual solvent, the samples were heat treated according to the sequence: 80ºC (30min); 100ºC (60min); 120ºC (120min); 150ºC (240min), with rates of $2ºC.min^{-1}$. The microstructure of one sample can be observed in the scanning electron microscope (SEM) image in Fig. 1.

2.2. Experimental setup

Optical pumping was performed with a frequency doubled Q-switched Nd:YAG laser. Pulses with maximum pulse energy of 3 mJ (at 532nm) were generated with duration of 10 ns at a repetition rate of up to 5 Hz using a saturable absorber Q-switcher of Cr^{4+}: YAG with a transmission of 55%. Short pump pulses are necessary in order to not excite the triplet states of rhodamine with long decay life time [5], once this excitation is recognized as the main limiting factor to the performance of dye lasers.

Coherent emission from the RL is an interference phenomenon which occurs preferably in the backscattering direction [6]. In this way, the backscattered luminescence of the studied sample is separated from the pump beam through a dielectric beam splitter and then captured for the spectral and temporal analysis. The experimental setup can be seen in Fig. 2.

Temporal and spatial profiles of the random laser emission are taken and the energy of the pump pulse is measured simultaneously, while the laser is operated in “single shot” mode.

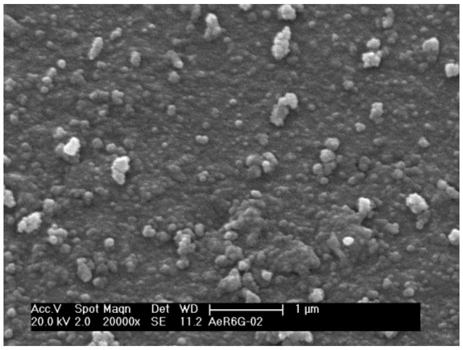

Fig. 1. SEM image of the rhodamine-doped aerogel under study.

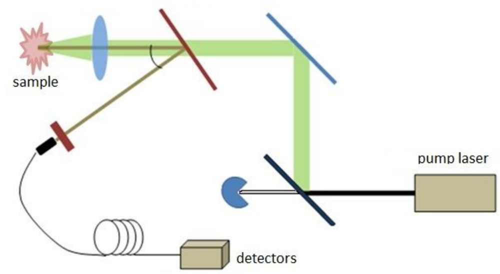

Fig. 2. Experimental setup.

3. Results and discussion

While increasing the energy of the pump pulse and measuring the temporal and spatial profiles of the random laser emission in the backscattered direction, the following can be observed: temporal and spectral narrowing, increased amplitude of the laser emission in the temporal and spectral profiles and higher RL pulse energy. Figures 3 and 4 show the temporal profile at low and high pump energies.

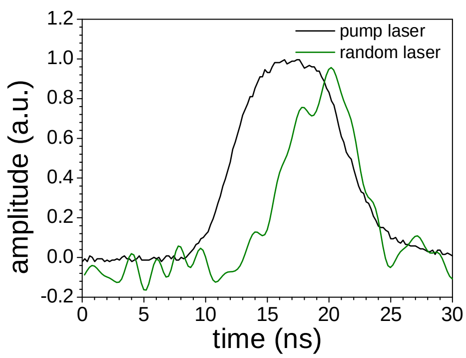

Fig. 3. : Temporal profile of pump pulse (pulse duration 9,7 ns - FWHM) and random laser (pulse duration 6,1 ns - FWHM) at low pump power.

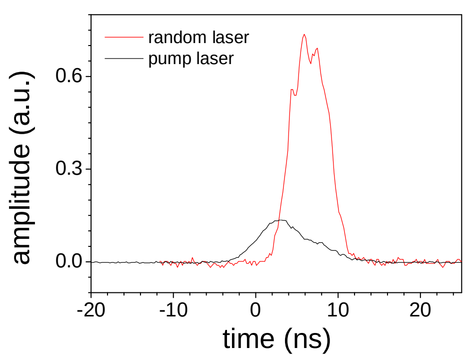

Fig. 4. Temporal profile of pump pulse and random laser obtained simultaneously with picoseconds detector at high pump power.

The spiky features of the RL emission which are not correlated with the pump pulse can clearly be observed, as shown in Fig. 4. We believe that this characteristics of the temporal profile of the RL emission indicate relaxation oscillations. The increasing of the optical energy of the RL pulse can also be clearly observed in Fig. 4.

The spectral features are demonstrated in Fig. 5 and 6. Figure 5 shows that the luminescence peak of rhodamine 6G in a silica matrix is shifted towards shorter wavelengths with respect to the luminescence peak in ethylene glycol. Spectral narrowing of the RL emission is shown in Fig. 6.

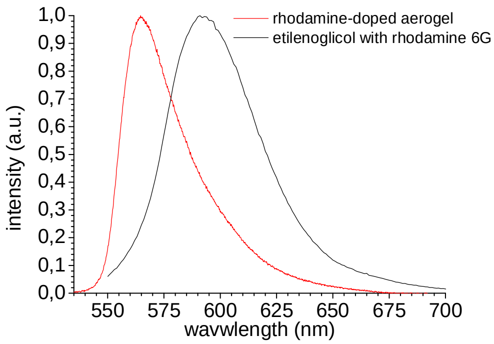

Fig. 5. Luminescence obtained for two different matrices doped with rhodamine 6G.

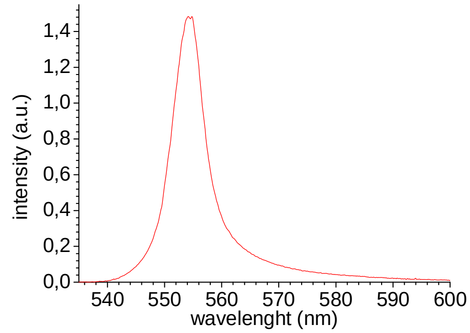

Fig. 6. Spectral profile of the random laser obtained with pump energy of 362 μJ.

4. Conclusion

We demonstrate for the first time random laser action from rhodamine-doped aerogel. By increasing the pump pulse energy we achieve spatial and temporal narrowing and increase of the emission energy of the random laser.

5. Acknowledgments

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6. References

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