Rhodamine-doped Aerogel Random Laser

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Abstract: Random Laser generation can give valuable information on the structure of diffuse materials. The aerogel matrix has knowingly a fractal organization. Here, random laser generation from an aerogel matrix doped with rhodamine 6G was established.

OCIS codes: (140.0140) Lasers and laser optics; (140.3380) Laser materials; (160.4670) Optical materials

1. Introduction

Silica aerogel is a material well known for its good mechanical and thermal properties. However, its optical properties have received much less attention [1], because it is optically not transparent. For the same reason, 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 lasers. The temporal and spectral characteristics are being studied as well as specific characteristics of scattering in aerogel matrix that need new approaches in order to be understood [2]. Moreover, the microstructure characteristics of the aerogel matrix can be controlled. The microstructure control is of great interest for the development and comprehension of the RL phenomenon [3], because the optical signal is strongly related to the structure of the scattering material [4,5].

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 (hydrolysis and gelation) were prepared from TEOS, ethanol, HCl and NH_4OH. Before the gelation occurred, the sol was transferred to polystyrene molds (1x1x4 cm), 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 and subjected to a heat treatment for remove any residual solvent.

2.2. Experimental setup

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. Optical pumping was initially performed with a frequency doubled Q-switched Nd:YAG laser. Pulses with maximum pulse energy of 3 mJ (at 532 nm) were generated with duration of 10 ns at a repetition rate of up to 5 Hz. Currently, all data have been reproduced using an Optical Parametric Oscillator (OPO) with pulses of 9 ns and 10 Hz. Temporal and spectral 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.

3. Results and discussion

In a first experiment, the luminescence profile of the rhodamine-doped aerogel was measured. The rhodamine luminescence peak in silica matrix occurs between 555 nm and 565 nm, which is shorter when compared to the luminescence peak in ethylene glycol. Figure 1 shows spontaneous emission at 555 nm of the rhodamine-doped aerogel sample during pumping at 532 nm.

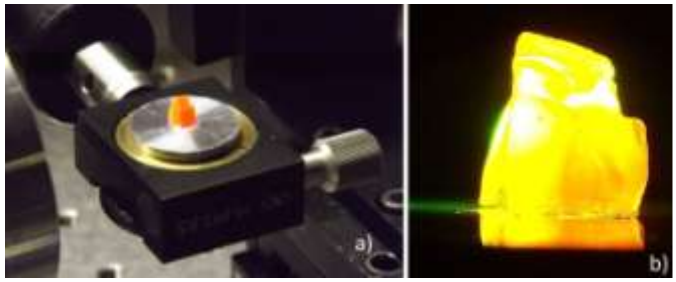

Fig.1: (a) Rhodamine-doped aerogel sample in setup, (b) scattered emission of the sample.

Increasing of the pump pulse energy generated a narrowing of the temporal and spectral profile of the RL emission in the backscattered direction, together with increased amplitude and higher RL pulse energy. In figure 2 the spectral narrowing and increase of the RL amplitude is shown. Besides the increase in pulse amplitude, emergence of several peaks is a feature of the RL emission in rhodamine-doped aerogel. Pump pulses of the order of 2.2 mJ generated 2 peaks (Fig. 2) and increasing the pump pulse energy causes the appearance of even more peaks. These peaks are very stable except for transition between two and three peaks. Therefore, the spectral profile goes through an instability before a emergence of a new peak with increasing pump power.

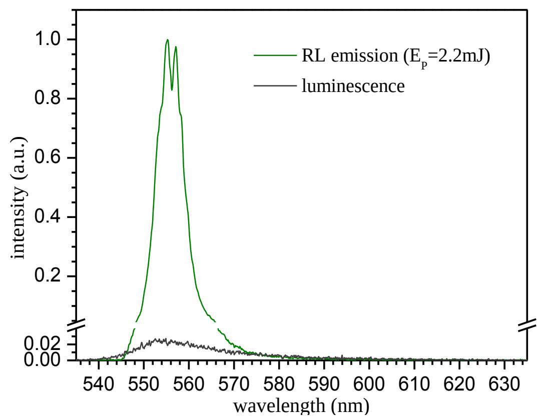

Fig. 2: Spectral profile of the random laser obtained with a pump energy of 2.2 mJ (green) and spontaneous emission in backscattered direction (gray) of the rhodamine-doped aerogel.

The characteristics seen before causes a difficulty to defining the transition between laser emission and spontaneous emission. However, analyzing the integrated output intensity as a function of pump energy shows clearly the typical behavior of a laser threshold (Fig. 3).

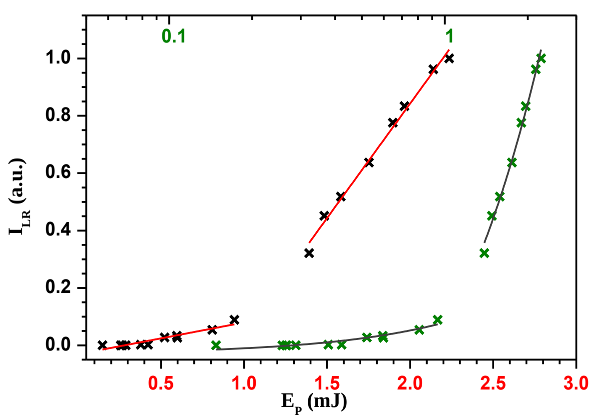

Fig. 3: Integrated output intensity (I_{LR}) as a function of pump energy (E_P) – linear scale (black) and logarithm scale (green).

The RL emission regime can be checked by linewidth analysis. Avoiding the instability interval, the linewidth as a function of pump energy shows the characteristic linewidth narrowing of the RL emission as seen in figure 3.

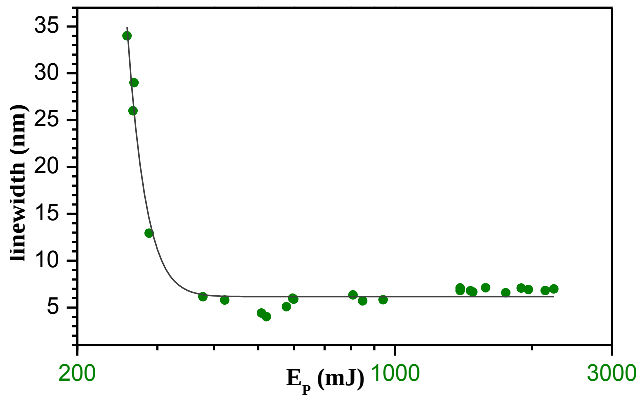

Fig. 4: Linewidth of the RL emission as a function of pump energy (E_P).

4. Conclusion

We demonstrate for the first time random laser action from rhodamine-doped aerogel. By increasing the pump pulse energy we achieve spatial narrowing and increase of the stimulated emission energy of the random laser. The RL emission features several peaks that will be related in a future publication to the structure of the rhodamine-doped aerogel under study.

5. Acknowledgments

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6. References

[1] W.J. Platzer, M. Bergkvist, "Bulk and surface light scattering from transparent silica aerogel", Sol. Energ. Mat. Sol. C. **31,** 243-251 (1993).

[2] X. Wu and H. Cao, "Statistical studies of random-lasing modes and amplified spontaneous-emission spikes in weakly scattering systems", Phys. Rev. A **77**, 013832-1–10 (2008).

[3] P. D. García,1, R. Sapienza, C. Toninelli, C. López, and D. S. Wiersma, "Photonic crystals with controlled disorder", Phys. Rev. A **84**, 023813-1–7 (2011).

[4] R.C. Polson, Z.V. Vanderny, "Random lasing in human tissues", Appl. Phys. Lett. **85,** 1289-1291 (2004).

[5] Q. Song, S. Xiao, Z. Xu, J. Liu, X. Sun, V. Drachev, V.M. Shalaev, O. Akkus, Y.L. Kim, "Random lasing in bone tissue", Opt. Lett. **35,** 1425-1427 (2010).

[6] X. Wu, W. Fang, A. Yamilov, A. A. Chabanov, A. A. Asatryan, L. C. Botten, H. Cao, "Random lasing in weakly scattering systems", Phys. Rev. A **74**, 053812-1–11 (2006).