Random Laser and Replica Symmetry Breaking in SiO_2-Rhodamine 6G xerogel powder

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Abstract. Random laser (RL) based on Rhodamine 6G (Rh6G) doped silica xerogel, fabricated by a conventional sol-gel (SG) synthesis, was observed around 590 nm, in a large band typical from dye RLs. Different from other previous works, where the xerogel is just impregnated or infiltrated with a dye solution, here the Rh6G was added during the SG synthesis. The obtained material was grinded using a mortar and a pestle, and the resulting powder was carefully packed in a sample holder and pumped at 532 nm using a 6 ns pulsed laser. We used spectral and images measurements to perform statistical analysis and describe experimentally the Parisi replica breaking symmetry (RSB) phenomenon in a complex system. These results show that RSB obtained from image is a promising method for RL characterization. Indeed, by calculating the RSB maps, we demonstrate that the RL emission is not a homogenous process, depending on the scattering and gain properties of different regions.

1 Introdution

Contrary to the conventional laser, where the optical feedback is performed by a defined cavity, random laser (RL), initially proposed by Letokhov [1], is a light source based on random and multiple light scattering inside a disordered medium, that provides the necessary feedback to achieve stimulated emission. Thus, RL emission properties depend strongly on the features of the disordered medium, turning it a good instrument for the study of materials properties.

In the present work, we report a broadband RL emission centered at 590 nm, observed from Rhodamine 6G (Rh6G) doped silica xerogel powder fabricated by a conventional sol-gel (SG) synthesis at room temperature. Spectral and images analyses are used for demonstrating replica symmetry breaking (RSB) phenomenon, one of the most relevant theories in statistical mechanics formulated in 1980' by the Nobel Prize Giorgio Parisi. In addition, we show that RSB is recognizably an important tool for RL characterization [2,3].

2 Methods

Initially, a 10^{-3} M Rh6G ethanolic solution was prepared. SG synthesis was carried out at room temperature with the addition of tetraethyl orthosilicate (TEOS), deionized water and nitric acid to the Rh6G solution, in the proportion of 1:1:1:0.1 % vol., respectively. The incorporation of Rh6G in the synthesis results in a more homogeneous distribution of the dopant in the sample. The solution was stirred during 15 min and, afterwards, the resulting sol was submitted to gelation and ageing at 50ºC during 72 h. The resulting xerogel was ground using a mortar and pestle to obtain a homogenous powder, that was carefully deposited on a microscope slide in a sample chamber with a thickness of 250 mm and was excited at 532 nm using a 6 ns Nd-YAG Q-switched pulsed laser.

Differently from conventional analyses, the RL threshold can also be defined statistically by considering the probability of the occurrence of RL emission. In this sense, some works have been performed to verify the occurrence of RSB phenomenon in RL [2,3]. This method consists of identifying a photonic phase-transition by measuring the degree of correlation between intensity fluctuations in different replicas (output spectra or images) but in the same experimental conditions.

For the RSB characterizations, we consider two different situations: for spectra analyses, the intensity fluctuation was measured for each mode (wavelength) i, and for images analyses the intensity fluctuation was measured for each pixel i, given by:

$$\Delta_\alpha(i) = I_\alpha(i) - \langle I\rangle(i) \qquad (1)$$

where $\langle I\rangle(i)$ is the average intensity for each mode (or pixel) over all measured replicas (100 for each excitation condition).

The overlap parameter $q_{\alpha\beta}$ between replicas α and β, is defined as

$$q_{\alpha\beta} = \frac{\sum_{i=1}^{N} \Delta_\alpha(i)\Delta_\beta(i)}{\sqrt{\sum_{i=1}^{N} \Delta_\alpha^2(i) \sum_{i=1}^{N} \Delta_\beta^2(i)}} \qquad (2)$$

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where N is the number of acquired spectral wavelengths (or pixel) points. This parameter is the photonic analogous to the Parisi overlap for magnetic system in the spin glass theory [4]. From the measured spectra (or images), we calculate the set of values of q to obtain the probability distribution $P(q)$ for each pumping energy, as in reference ref. [3].

3 Results and Discussion

In Fig. 1 we illustrate the $P(q)$ distribution using the values of q, calculated from Eq. 2 from 100 of images (100 of replicas). At low pumping energy, most of the q values are centered around zero, meaning that the emitted modes are independent and do not interact or interact weakly with each other, Fig. 1(a). As the energy increases, the modes are coupled by the nonlinearity and q assumes values close to -1 or 1, Fig. 1(b). Such behavior characterizes the RSB phenomenon and can be used to identify the transition from spontaneous to RL emission.

Fig. 1(c) shows the $|q_{max}|$ for images (blue curve) and for spectra (red curve) as a function of the pumping energy. They present similar behavior despite a small shift in the energy where the RSB transition occur: 0.40 mJ and 0.60 mJ, respectively. The reducing of the full width at half maximum (FWHM) of the emission band (black curve) from about 50 nm at low pump energy to about 25 nm at high pump energy is another evidence of RL emission and can be attributed to the fact that the increase of the number of photons at the maximum gain frequency is faster than the increase at other frequencies, due to the resonant properties of the scattering medium.

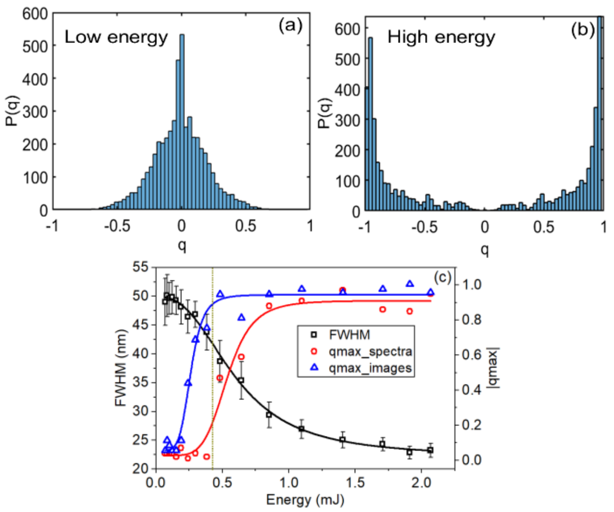

Fig. 1. *P(q)* distribution at (a) low energy and (b) high energy from 100 of images. (c) FWHM and RSB characterization as a function of the pumping energy.

Differently from spectral analysis, where it is possible to analyze the excited area only as a whole, by using images we can analyze the spatial distribution of the emission modes. Fig. 2(a) represents the mean intensity image obtained from the set of 100 images obtained from the sample excited at 1 mJ (above RL threshold). We can see that regions under the same excitation energy have different emission intensities. In our work, we divided each image in 256 sub images in order to obtain 16x16 image grid. Then the set of q values was calculated for each sub image and the most frequent value of |q| ($|q_{max}|$) was determined for each cell in the image grid. Fig. 2(b) shows the $|q_{max}|$ map obtained from Fig. 2(a), under the same pumping condition. The results indicate that correlating fluctuations and RSB phenomenon is not homogenous in the sample. Some points of the material are RL switched ON (yellow spots) and others are OFF (blue spots), depending on the scattering and gain properties of each region.

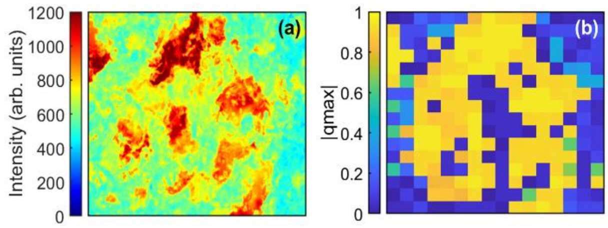

Fig. 2. (a) Mean intensity image pumped at 1 mJ and (b) Respective map with the mostly frequent value of $|q|$ ($|q_{max}|$).

4 Conclusions

A Rh6G doped silica xerogel was successfully synthesized for the purpose of RL emission spectra and images analyses. We observed RSB phenomenon with the analogous Parisi overlap parameter changing from 0 to ± 1 for excitations above the RL threshold. This phenomenon is attributed to RL emission. The optical feedback responsible for the laser action is attributed to light scattering due to reflections between the powder particles. Complementary, image results also demonstrated that the RL emission is not a homogenous process, indicating that some points of the material achieve RL before others, depending on the scattering and gain properties of different regions. Therefore, emission image and RSB mapping is a promising method for RL characterization.

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