Angular distribution of random laser emission

F. Tenopala-Carmona,[1] C. García-Segundo,[1,*] Natanael Cuando-Espitia,[2] and Juan Hernández-Cordero[2]

[1]*Centro de Ciencias Aplicadas y Desarrollo Tecnológico, Universidad Nacional Autónoma de México, Circuito Exterior S/N, Ciudad Universitaria, AP 70-186, CP 04510, México D. F., Mexico*

[2]*Instituto de Investigaciones en Materiales, Universidad Nacional Autónoma de México, Circuito Exterior S/N, Ciudad Universitaria, AP 70-360, CP 04510, México D. F., Mexico*

**Corresponding author: crescencio.garcia@ccadet.unam.mx*

Received October 31, 2013; revised December 18, 2013; accepted December 25, 2013; posted January 2, 2014 (Doc. ID 200214); published January 29, 2014

We present an experimental analysis of the angular distribution of the emission of a random laser (RL) operating in the diffusive regime (~1% volume fraction of scatterers). The RL ensemble was made of silica particles suspended in a 1:1 methanol:water matrix with Rhodamine 6G dye as the active medium. We found that, for specific pumping-energy-dependent scattering strength, the RL spectrum reached stable features that were angularly preserved. From the analysis, we propose that this defines a novel parametric condition that may well be the equivalent of the RL critical volume, as proposed by Letokhov.

OCIS codes: (140.2050) Dye lasers; (290.4210) Multiple scattering; (160.3380) Laser materials; (160.6030) Silica; (260.2710) Inhomogeneous optical media.

http://dx.doi.org/10.1364/OL.39.000655

Random laser (RL) systems have become a fast growing topic of interest owing to their scientific and technological relevance [1]. An important aspect in the study of these light sources is to establish the parametric conditions of operation, which are mainly related to the disposition of the scattering ensemble within the gain medium. In RL systems, multiple scattering promotes long optical path lengths (OPLs) leading to the narrowing of the emission spectrum. Since the path length depends on the scattering strength, one can vary the volume fraction of scatterers and analyze the spectral narrowing over long OPLs and then relate them to the cascade of stimulated emission events [2,3]. As lasing is restricted to wavelengths whose stimulated emission efficiency can be assured along large enough optical paths, the favored spectral emission is required to travel much longer OPLs. Moreover, as this process involves a certain scattering volume, including amplifying (excited) and absorbing (unexcited) regions [1,4], then it must be considered that lasing intensity tends to increase through the excited volume while it is damped across the unexcited one. The extended scattering strength is therefore expected to modulate the intensity, the spectral properties, and the spatial distribution of the dominant amplified extended modes. In this context, in a 3D-thick random medium (3DRM), the analysis of the spatial distribution of the lasing emission exhibits specific spectral features that shall depend upon the point of observation.

In the random medium with gain, the interdependence between the scattering and gain parameters, and its effects on the RL output is not so evident. In general, the scattering strength interplays with the optical gain and the gain saturation to yield RL emission. While the width of the optical gain is determined by the amplifying medium, the bandwidth of the RL is determined by the scattering characteristics and by the photon density of the external pumping source. Therefore, the feasibility for achieving laser emission depends on the interplay between the concentration of scatterers (assumed as nonabsorbing), the concentration of the gain molecules (optical absorption distribution), and the decay efficiency of the gain medium. When the lasing conditions are reached, the photon density distribution of the decaying process would experience spectral narrowing resulting from the traversed OPLs (i.e., the scattering strength) [2]. It is commonly assumed that, for a large enough photon distribution and long enough average OPL, the optical output features would tend to converge to a stable laser mode [2]. For instance, it has been shown in [5] that the increase of the concentration of scatterers yields both an enhancement of the laser intensity and a narrowing of the spectrum. With this in mind, our point of interest is to determine whether the conditions for obtaining a stable laser mode can be achieved for any point of observation and how this depends on the scattering strength. In other words, we aim to analyze the dependence of the optical output on the particle concentration as well as on the point of observation, i.e., the angular variations of the RL emission.

In this study, we analyzed phenomenologically the spatial and spectral features in a RL system where the colloid volume (i.e., the 3DRM) was larger than the excitation volume. Thus, we focused on the experimental analysis of the content of information in the RL emission and on its angular distribution. We believe this is relevant since in the literature, several conclusions are drawn regardless of appropriate considerations of the point of observation of the spatial distribution of the light emerging from the scattering volume. Similarly, the fact that not all the 3DRM is excited is seldom considered in analyzing the spectral emission of RLs. There is experimental evidence showing that the spectral features of RLs depend on the angle of observation [6–8]; however, these studies have been performed on the localized regime, i.e., for large scattering strengths [1]. In our experiments, the RL system operates in the diffusive regime, leading to substantially different results than those obtained for RLs in the localized regime. Furthermore, in contrast to previous work based on rectangular configurations [6–8], our analysis of the spatial and spectral characteristics of the RL emission is based on the use of a cylindrical geometry. The aim was to identify, experimentally, the

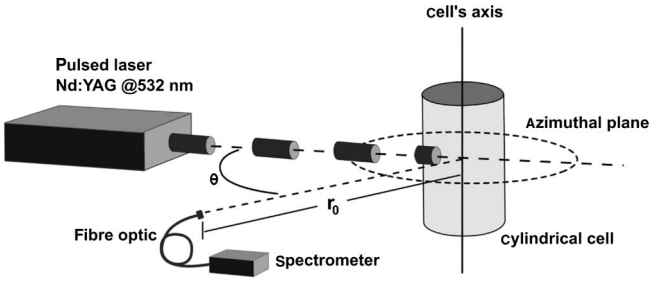

Fig. 1. Schematic of the experimental array. The input end of the optical fiber is mounted on a rotatable arm yielding angular displacement (θ) around the cylindrical cell (10 mm diameter). The pump beam defines the azimuthal origin (i.e., $\theta = 0$) around the cell.

actual parametric conditions for achieving random lasing with stable and maximized intensity over a given spatial distribution, regardless of the point of observation.

Our experiment was designed to register the optical output for two different ensembles: a scattering colloidal sample and a RL system. For both cases, we studied the optical output as a function of the excitation energy and particle concentration (otherwise volume fraction). Hence, we performed measurements at various angles using an optical fiber mounted on a rotatable arm pinned to match the axis of a cylindrical glass cell, as depicted in Fig. 1. The input end of the fiber was placed at a fixed distance away from the surface of the cell ($r_0 = 28$ cm) and the other end was connected to a high resolution spectrometer (Ocean Optics HR4000). The samples were allocated in the cell, which was held slightly tilted with respect to the pump laser beam in order to prevent back reflections from the surface. For all experiments, the sample was pumped with a pulsed laser impinging on the center of the cell. The pumping laser (Quantel, Mod. Brio) was a pulsed Nd:YAG set at 532 nm with a 10 Hz repetition rate and temporal pulse width of 7 ns with a constant spot diameter of 4 mm. For data analysis, we defined the pump beam to be the azimuthal origin around the cylindrical cell.

Because of the symmetry of the experimental configuration, we chose to register the optical output at angles ranging from 20° to 160° in steps of 10°. Due to strong saturation of the spectrometer, we avoided measurements for angles smaller than 20° and angles within the range of 160°–180°. The pump energy range was set between 4.2 and 29.6 mJ per pulse. This range of pump powers has been previously shown to yield RL emission in silica-based RL [9]. From the registered spectra we obtained the intensity distribution of the scattered light as function of the pump energy and of the angle of observation. For the case of RL samples we obtained the peak wavelength of the emission, the full width at half-maximum (FWHM), and the evolution of the spectra while varying experimental parameters, such as pump power, particle concentration (scattering strength), and angle of observation.

Since the scattering phenomenon is at the core of the lasing process, we first analyzed the intensity distribution of the pump light scattered by the particles. In this case, the colloid sample was made of SiO_2 spherical particles of 200 nm ±16% in diameter, which were grown by the modified Stöber method [10]; this choice was made because these particles have low optical absorbance within the range of interest [11]. This particle size was selected because the coherent reflectance in a colloid with spherical particles is maximized when the particle diameter is around half the incident wavelength [12]. In our scattering experiments, the particles were suspended in a matrix of 1:1 bi-distilled water and methanol at concentrations of: 1.05×10^{11}, 2.5×10^{11}, 6.30×10^{11}, 1.26×10^{12}, 2.63×10^{12}, and 3.98×10^{12} particles per milliliter. Thus, our experiments were conducted in the diffusive regime [1]. The RL experiments were conducted using the same type of colloids as before, but this time Rhodamine 6G was added at a concentration of 2.5×10^{-3} M, following the criteria given in [13]. Intuitively, one can assume that as long as the experimental conditions remain in the diffusive regime, the physics of the phenomenon remains the same, as is the case in all of our experiments.

The intensity of the light scattered by the colloidal sample as function of observation angle is shown in Fig. 2(a). As the particle concentration increases, the backscattering also increases and a lesser portion of light traverses the sample along the forward direction. In this case, the number of potential scattering events also increases [14], as does the average OPL. We also observed that for a given particle concentration, the intensity distribution scales with the pump energy. This means that absorption or other nonlinear effects were negligible for these experiments [13].

The intensity of the RL emission as function of angle of observation is plotted in Fig. 2(b). Clearly, most of the optical output exits the RL ensemble in the backward direction. The decrease in the intensity at larger angles implies that the light travels a longer OPL along the unexcited volume. This means that the magnitude of coherent amplification achieved in the RL ensemble is linked to a large enough OPL within the excited volume as well as to a small OPL within the unexcited region [15]. Notice that the angular variation of RL emission was less concentration-dependent than the scattering intensity. As the particle concentration increases, the RL intensity also increases and tends to saturate for the largest concentrations.

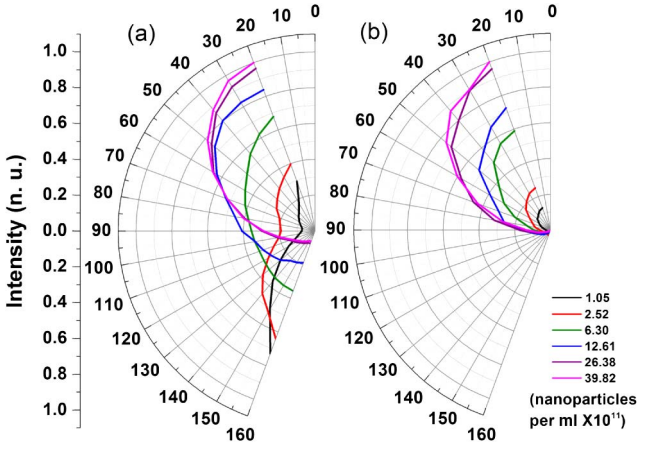

Fig. 2. Angular measurements for different particle concentrations: (a) scattering of the pump beam from the particles and (b) intensity of the RL emission.

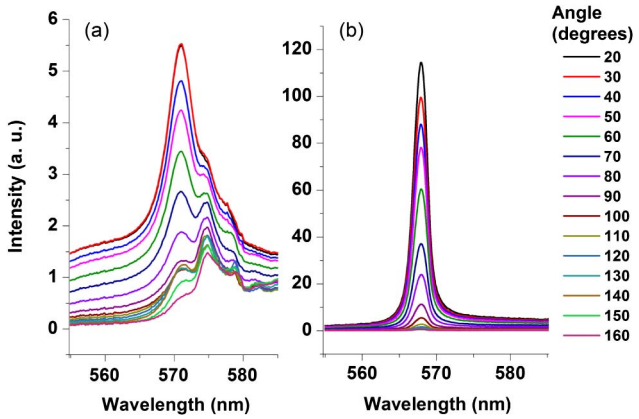

Fig. 3. RL spectrum for different angles and for a fixed pump energy (5 mJ/pulse): (a) 1.05×10^{11} particles per milliliter and (b) 3.98×10^{12} particles per milliliter.

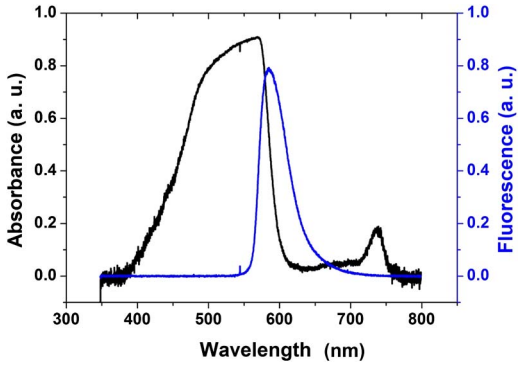

Fig. 4. Absorption and fluorescence spectra of the gain medium (Rhodamine 6G).

Figure 3 shows the spectral evolution for different observation angles, a fixed pump energy, and two different particle concentrations. For a concentration of 1.05×10^{11} particles per ml, as the observation angle varied, the spectral emission of the RL varied as shown in Fig. 3(a). In contrast, as seen in Fig. 3(b), for a higher concentration of particles (3.98×10^{12} per ml) we obtained stable features in the spectrum for different angles. This represents a condition of particle concentration and pump energy for which the spectral features of the RL are preserved, regardless of the angle of observation. This suggests that for these experimental conditions, the extended modes supported by the RL ensemble were uniformly amplified in the excitation volume. The existence of such modes has been predicted theoretically and numerically in previous reports [1,2]. As a backup to this assumption, we further observed that the increase in the scattering strength of the system always reduced the FWHM of the laser peak yielding a 'selection' of the amplified modes. Furthermore, we consistently observed that, for every particular pumping condition, there was a certain 'threshold' for particle concentration that yielded stable wavelength emission in all directions. In these cases, an increase in the pump energy leads only to an increase in the FWHM of the RL peak. Below this threshold of particle concentration, the RL ensemble did not yield angularly stable spectra; regardless of the pump power.

Based on our experimental conditions, we set our discussion on a diffusive model as proposed in [3,15]. The pumping beam entering the RL ensemble is diffused and decays in intensity according to Lambert–Beer's law. In the gain medium, an initial density of excited molecules is set, and its spatial distribution is a function of the intensity of the pumping beam within the sample. It is important to remark that because of the exponential decay of the intensity, one can differentiate between two main zones: one with a high density of excited molecules and the other with a low density, henceforth labeled as the excited and the unexcited regions, respectively. After a given time τ, a certain amount of molecules decay by means of spontaneous emission. The resulting emitted photons travel along random paths, either generating stimulated emission (as long as they are in presence of excited dye molecules), or being absorbed if they are in presence of unexcited molecules. Therefore, the initial spontaneous emission will be amplified as long as it travels across the excited region, whereas it will be absorbed while traversing the unexcited region.

Different wavelengths are neither equally absorbed nor equally emitted by the dye molecules (see Fig. 4 showing the absorption and emission spectra of the Rhodamine 6G dye), and the inhomogeneous absorption of the emission spectrum might produce inhomogeneous quenching [Figure 3(a)]. Therefore, one must either reduce the quenching or narrow the spectrum in order to get homogeneous spectral features along the whole angular scan.

According to the stimulated emission phenomenon, another effect that must be observed while increasing the OPL across the excited region is the narrowing of the emission spectrum. As can be seen from Fig. 2, a higher concentration of nanoparticles enhances the scattering of the pumping beam. A stronger scattering process yields two main consequences: a larger fraction of the pumping beam is absorbed by the gain medium, and the number of scattering events is also increased. A higher absorption of the pump beam implies a higher number of excited molecules and therefore an increase of the excited volume. An enhanced scattering strength implies larger average OPLs for both the pumping beam and the RL emission. Therefore, the increase in the scattering strength not only reduces the inhomogeneous quenching by decreasing the number of absorbing molecules, but it also narrows the RL spectrum by increasing the average OPL within the system.

The above phenomenon is directly exhibited in Fig. 3, and it shows the direct interplay between gain and OPL driven by changes in the concentration of scatterers. Specifically, it shows that the latter is a parameter that increases both gain and OPL. Furthermore, based on our finding of a pump-dependent threshold concentration of scatterers at which the spectral parameters (peak wavelength and FWHM) remain constant along the whole angular sweep, we can infer that this represents the condition at which gain saturation is reached. In other words, at this concentration threshold, the light travels within an OPL with gain which is long enough to yield stable spectral conditions providing a narrow spectrum in every direction.

We have stated that this threshold concentration is pump-dependent, and also that the only influence of the pump on the stable spectral features is an increase in the FWHM. This can be explained by means of conservation of energy in the following fashion: a higher number of pumping photons in a particular scattering ensemble increases the number of excited molecules without increasing the average OPL. The probability of re-absorption and re-emission events is therefore increased yielding photons at longer or equal wavelengths according to conservation of energy, thereby increasing the FWHM. It has also been shown that mode coupling can occur in similar RL systems and this can also affect the FWHM [16].

Relating the concentration threshold with the three parameters mentioned by Wiersma in [2] (gain, gain saturation, and mode lifetime), we can say that we modulated the interplay between gain and mode lifetime (related to the OPL) to the point at which gain saturation in a certain 3DRM volume was effectively achieved. This volume represents the same critical volume as that proposed by Letokhov in [4]. Note that due to the relation between the concentration of scatterers and the OPL, the higher the concentration, the smaller the sample's volume required to get stable spectral RL features and vice-versa. Additionally, the smaller the critical volume, the lower the pump energy required to excite the active molecules. Therefore, our three experimental parameters, namely concentration of scatterers, pump energy, and an optimized concentration of active molecules, play the same role as the gain, gain saturation, and mode lifetime.

Finally, we propose that the method reported here, with the additional use of careful calculations of mean free paths and scattering coefficients, can be useful for experimentally measuring the actual critical volume of a RL system as a function of the concentration of scatterers and of the pump energy, while the size of the scatterers and the density of active molecules are kept constant.

We thank R. Sato-Berrú and J. Flores-Flores for providing the SiO_2 particles. This work was partially funded by CONACyT (grant 154464), DGAPA-UNAM (grant PAPIIT-IN102112), and by a grant contract between CCADET-UNAM and the ICyTDF in Mexico City.

References

1. D. S. Wiersma, Nat. Phys. **4**, 359 (2008).
2. D. S. Wiersma, Nat. Photonics **3**, 246 (2009).
3. S. Mujumdar, M. Ricci, R. Torre, and D. S. Wiersma, Phys. Rev. Lett. **93**, 053903 (2004).
4. V. S. Letokhov, Zh. Eksp. Teor. Fiz. **53**, 1442 (1967).
5. A. M. Brito-Silva, A. Galembeck, A. S. L. Gomes, A. J. Jesus-Silva, and C. B. de Araújo, J. Appl. Phys. **108**, 033508 (2010).
6. R. K. Thareja and A. Mitra, Appl. Phys. B **71**, 181 (2000).
7. S. F. Yu, C. Yuen, S. P. Lau, W. I. Park, and G.-C. Yi, Appl. Phys. Lett. **84**, 3241 (2004).
8. H. Cao, Y. G. Zhao, H. C. Ong, and R. P. H. Chang, Phys. Rev. B **59**, 15107 (1999).
9. N. Cuando-Espitia, J. Hernández-Cordero, C. García-Segundo, and R. Quispe-Sicha, in *Advanced Photonics* (Optical Society of America, 2011), paper IMD5.
10. R. Sato-Berrú, J. M. Saniger, J. Flores-Flores, and M. Sanchez-Espíndola, Mater. Sci. Eng. **3**, 237 (2013).
11. I. S. Altman, D. Lee, J. D. Chung, J. Song, and M. Choi, Phys. Rev. B **63**, 161402 (2001).
12. R. G. Barrera and A. García-Valenzuela, J. Opt. Soc. Am. A **20**, 296 (2003).
13. M. Villagrn-Muniz, C. Garcia-Segundo, H. F. Ranea-Sandoval, C. Gogorza, and G. M. Bilmes, Appl. Phys. B **61**, 361 (1995).
14. T. Mokhtari, C. M. Sorensen, and A. Chakrabarti, Appl. Opt. **44**, 7858 (2005).
15. D. S. Wiersma and A. Lagendijk, Phys. Rev. E **54**, 4256 (1996).
16. H. Cao, X. Jiang, Y. Ling, J. Y. Xu, and C. M. Soukoulis, Phys. Rev. B **67**, 161101 (2003).