Diffusive random laser modes under a spatiotemporal scope

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Abstract: At present the prediction and characterization of the emission output of a diffusive random laser remains a challenge, despite the variety of investigated materials and theoretical interpretations given up to now. Here, a new mode selection method, based on spatial filtering and ultrafast detection, which allows to separate individual lasing modes and follow their temporal evolution is presented. In particular, the work explores the random laser behavior of a ground powder of an organic-inorganic hybrid compound based on Rhodamine B incorporated into a di-ureasil host. The experimental approach gives direct access to the mode structure and dynamics, shows clear modal relaxation oscillations, and illustrates the lasing modes stochastic behavior of this diffusive scattering system. The effect of the excitation energy on its modal density is also investigated. Finally, imaging measurements reveal the dominant role of diffusion over amplification processes in this kind of unconventional lasers.

OCIS codes: (140.3460) Lasers; (140.3380) Laser materials; (290.4210) Multiple scattering; (300.6500) Spectroscopy, time-resolved.

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1. Introduction

Random lasers (RLs) are open sources of stimulated emission whose feedback is provided by disorder-induced light scattering mediated by random fluctuations of the dielectric constant in space. Among them, those in which light undergoes diffusive motion within the gain medium, the so-called diffusive random lasers (DRLs), are of particular interest since a clear modal picture of such extremely leaky systems is still missing in the literature. Here, the complexity arises from the strong nonlinear interaction between both spatially and spectrally overlapping lasing modes. As with most lasers, in a DRL, lasing occurs above a pumping power threshold. However, pump and emitted photons are involved in multiple scattering processes which increase the dwell time of photons inside the material and allow for enough amplification to compensate for absorption and leakage of light through its boundaries creating gain saturation. A classical DRL is usually realized by closely assembled optically active particles (such as laser crystal or amorphous powders) that scatter light and have gain, or by passive scatterers embedded in an amplifying medium. Note that random lasing in diffusive systems with a continuum spectrum was the first observed and theorized. Early RL experiments in colloidal dye solutions showed above threshold, an overall spectral narrowing and a rapid increase of emission intensity at the frequency of maximum gain [1]. In such a case, the emission dynamics can be described by a diffusion equation with gain, where the phase of the light field, and hence interference, do not play any role [2, 3]. This laser radiation, which is not dominated by frequencies of a resonator, has been called lasing with nonresonant feedback. In contrast to the mentioned smooth and relatively broad lasing spectra, later experiments by Cao and associates revealed the presence of multiple narrow spikes in the emission spectrum of zinc oxide powders [4]. As for ordinary lasers, light emitted at these peak frequencies has a Poissonian photon statistics which demonstrates the temporal coherent nature of this emission [5]. Originally, it was suggested that in the regime of localization, recurrent light scattering events could be behind these spectrally narrow emission features as they could serve as ring cavities for light providing resonant feedback [6–8]. However, more recent experiments showed that narrow emission spikes arise in nearly the entire range of accessible scattering strengths, *i.e.* for strong, diffusive, and weak scattering [9–11]. The key parameters discerning among the three mentioned regimes are the transport mean-free path l_t, over which the direction of light propagation is randomized, the size of the sample L, and the wavelength of the light [12]. Apalkov *et al.* suggested that in the diffusive regime ($\lambda << l_t << L$), ring-shaped resonators with a larger refractive index than average might have some nonnegligible probability to exist. Random lasing studies in π-conjugated polymers supported this model [13, 14]. However, these configurations can only exist if spatial fluctuations of the refractive index are correlated over large enough distances, so they will not appear in all weakly scattering cases (see [15]). Mujumdar and associates proposed another scenario. They suggested that among the spontaneously emitted photons of a random system with gain, there exists a subset of rare photons that travel much longer distances than the average ones. These 'lucky photons' accumulate enough gain to activate a new lasing mode with a different wavelength after each excitation shot, and give rise to random spikes in the spectrum [10, 16]. The experimental study performed by Chabanov *et al.* has brought forward the existence of long-lived extended modes in regular diffusive materials which might be responsible for the observed narrow spikes in RLs [17]. However, it was recently shown that strongly localized modes can co-exist with modes of much larger spatial extension which become more easily coupled from mode competition. Depending on the material studied, both localized and extended modes can thus lase and provide a coherent RL mechanism [18]. On the other hand, DRLs are highly multimode as many spatially overlapping modes exist, so mode mixing,

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which obscures the characteristics of individual modes and is likely to be modified by the boundary conditions, is particularly important. If the mode overlap is significant, complex nonlinear processes such as temporal oscillations [19, 20] or spatial hole burning [21] may appear. Gain mode competition may also play a crucial role when determining the condition of the different statistical regime of fluctuations and spectral profiles of RLs [22]. The intensity distribution of RLs obeys a Lévy type defined power-law tails [23]. However, different statistical regimes and crossovers are possible depending on gain, scattering strengths, excitation energies, and sample sizes [24–26]. Furthermore, the mode coupling between long-lived extended modes together with the stochastic behavior inherent in the spontaneous emission from which the RL starts at each shot, can explain the strongly stochastic behavior in the positions of the narrow emission lines of an RL with static disorder reported by Mujumdar *et al.* [16].

On the other hand, the presence of coherent multimode lasing even in the diffusive regime, has been a challenge for conventional laser theory. The focus of theoretical research in this field is to extend the semiclassical multimode lasing theory to open and irregular systems, the modes of which have a broad distribution of radiative lifetimes and an irregular spatial pattern [27]. As the usual methods for introduction of modes and quantization based on eigenvectors of hermitian operators are not applicable in such a case, alternative methods for defining electromagnetic modes are proposed by using the quasimodes, quasi-bound (QB) states, or resonances of the passive system without gain [27, 28]. In the strong scattering regime, the eigenvectors resulting in these theoretical explorations are nearly identical (within the scattering medium) to the threshold lasing modes. However, the difference between them increases for more lossy systems such as DRLs [28]. A recent time-independent theory based on the so-called constant-flux (CF) states which can find the random lasing modes and frequencies self-consistently (with no relation with the QB states), allows one to study the multimode regime in DRLs, and provides detailed information about the effects of mode competition through spatial hole burning [29–31]. Within this approach, uniformly spaced frequency spectra are expected in DRLs due to their modal interaction through the gain medium. In accordance with it, some experimental observations show more or less regularly spaced lasing frequencies exhibiting mode repulsion [15, 32, 33]. No significant changes in the lasing frequencies were either predicted for different pump strengths or spatial profiles of the pump whereas the intensities vary strongly. This behavior agrees with results presented in [33]. Nonetheless, the CF theory only improved the understanding of the properties of lasing modes in the stationary regime. RLs are intrinsically time-dependent systems due to the different time scale of transport and population dynamics, so in order to address the description of the dynamics, structure, frequency or intensity statistics of lasing modes in a DRL, a time-dependent model in three-dimensional (3D) RL structures, suitable under local and pulsed-pumping conditions, might be necessary. Bearing in mind the great computation effort required for this kind of numerical and theoretical explorations and puzzled by the diversity of interpretations proposed in the literature on the lasing mode nature, we decided to accomplish a thorough experimental study of the modal behavior of a representative diffusive random laser.

In this work, we explore the RL behavior of a ground powder of an organic-inorganic hybrid compound based on Rhodamine B (RhB) incorporated into a di-ureasil host. In a previous report, we investigated the excited state relaxation of the RhB molecules by using a time-resolved spectroscopy based on the RL generation of monomers and dimers [34]. In such a study, we were interested in the RL response of the dye in this solid state matrix under typical diffusive conditions. Therefore, we employed a broad pumped area and averaged hundreds of emission shots. It is worthy to notice that the observation of isolated narrow lines or a global narrowing of the RL spectrum often depends on the chosen experimental configuration. The ideal conditions for observation of spiky spectra are the use of a short-time pump pulse duration (~30 ps) and single-shot observation. Both the collection of several

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emission shots, or the use of a long enough single excitation pulse, may lead to an averaging mechanism among modes which reduces the previously mentioned fluctuations and gives rise to a smooth emission structure. This is probably the reason why some early RL experimental studies missed the observation of narrow spectral features in DRLs [35]. A reduction of the spikiness of the emission spectra, accompanied by an increase of the intermode spectral correlation, can be also achieved by adjusting the shape of the pump beam with a spatial mode modulator to incrementally excite larger numbers of spatially separated lasing modes [36]. The observed transition from a spiky to a smooth emission profile was explained by a phase locking between modes. On the contrary, the reduction of the spatial extent of the pump applied to the gain material by using a tightly focused beam, facilitates the experimental observation of well separated sharp peaks [37]. When a large enough pump spot is employed, narrow lasing lines overlap and the emission spectrum becomes smooth. In contrast, smaller pump areas excite fewer modes, so a fine structure can thus be observed [38]. On the other hand, recent results on the spatial structure of random laser modes can be found in [39–41].

Here, the aim is to give experimental evidences of the modal structure and modal oscillation dynamics of our diffusive scattering system after single shots of 30 ps pulse duration. As already pointed out, the difficulty in unscrambling the complex lasing behavior of a DRL is caused by the large number of modes that can simultaneously lase and randomly couple. As a result, the lasing modes dynamics of a DRL is rarely experimentally investigated [42, 43]. In almost all reported RL experiments with pulse laser excitation, time-integrated data are recorded. Under these circumstances, it is hard to extract any detailed information about individual lasing modes and thus impossible to know whether different modes coexist at the same time or appear subsequently. However, these difficulties can be avoided with the novel experimental arrangement adopted in this work. Our setup was specially designed to enable spatial mode selection, which favors the observation of individual lasing spikes, as well as accurate time-resolved measurements which permit to follow experimentally their dynamics response. The use of a streak camera allows a 2 ps time resolution which is a crucial requirement in this regard. The way to resolve lasing modes consists both in using a small enough pump spot size and in selecting a small enough emitting surface area by spatial filtering in the detection. The performed systematic study of the temporal evolution of the spectrally resolved emission evidences that different lasing modes stem from different time intervals showing a different temporal behavior in a DRL and illustrates the existence of relaxation oscillations from the time traces of individual lasing modes. In this approach, the modal density dependence on the pump energy and pump spot size is also experimentally accessed. Careful examination of the temporal and spectral response of the corresponding sharp lasing peaks allows us to assess the stability of the mode structure between different shots and, therefore, to evaluate the stochastic behavior of these active random media. The analysis of the results shows that our powder system lases in different modes in successive excitation events under constant experimental conditions. Parallel to this research, we have examined the spatial properties of the RL emission of this sample. We herein explore whether the imaging of the RL light on the dye doped powder surface is spatially restricted to the pumped domain (as suggested in the literature [37]), or else it is extended over a larger area of the scattering medium.

We believe that the outcome of our experiments will help the RL community to deepen the understanding of fundamental and intriguing aspects of DRLs such as their spatiotemporal behavior.

2. Experimental

2.1 Sample preparation and characterization

In this paper, we try to drive some definite conclusions about the lasing mode structure and modal oscillation dynamics of a ground powder sample of a d-U(600) di-ureasil hybrid

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containing 1.23×10^{19} RhB molecules/cm^3, which was prepared by the two-step sol-gel synthesis process presented elsewhere [44] and was ground by using a mixer mill. The polydispersity of the explored powder was evaluated from images obtained in a Confocal Microscope. The histogram of the particle size was fitted to a log-normal function. An average powder size of 6.6 μm was found. The resulting powder was compacted afterwards in a 6 mm high cylindrical quartz cell without a front window for handling ease. The volume filling factor of the powder material (f = 0.55) was calculated by measuring sample volume and weight. In a similar hybrid powder doped with 4.96×10^{18} RhB molecules/cm^3, we have experimentally estimated the transport mean free path, l_t, by studying the exponential decrease of the diffusive transmittance with the powder thickness. By fitting the experimental results to the theoretical modeling of transmittance given in [45], the obtained value for l_t in such a powder sample was 14 ± 2 μm at 630 nm, which confirms the diffusive scattering regime in the explored di-ureasil powder.

2.2 Experimental techniques

Time-resolved measurements were performed at room temperature by using the experimental setup shown in Fig. 1. The powder sample was excited by a frequency doubled mode-locked Nd: YAG laser (EKSPLA, PL2251B) with a pulse duration of 30 ps and 20 Hz. The laser excitation energy was attenuated with a half-wave plate combined with a polarizer and measured with an energy meter. A beam splitter (BS1) was employed to reflect about 5% of the pumping green laser to an optical delay line which provides a reference signal. The laser beam was focused to a spot size of 500 μm on the sample surface. The emission from the front face of the sample was collected with the L2 lens (f = 5 cm) placed at twice the focal length distance ($S_{L2} = 2f$) and imaged on a pinhole (P) positioned at the same distance ($S_P = S_{L2}$) behind the lens. As lateral magnification is one, this experimental arrangement allows selecting emission from an excited area equal to the employed spatial filter aperture controlling the number of the detected lasing modes. Note that pinholes of different diameters were used (Ø = 50, 30 and 15 μm). A long-pass filter (F) was employed to remove light coming from the sample at the pump frequency. A lens (L3) placed behind the pinhole focuses signals coming from the two optical paths on the entrance slit of an imaging spectrograph (Chromex C5680 Hamamatsu) connected to a streak camera (C5680 Hamamatsu). The lengths of both optical paths were previously made equal, so the barycenter of the pumping pulse was used as time reference. Both the RL pulse and the pumping pulse signal were simultaneously detected in single shot measurements. The time window of the streak camera was set at 1 ns. Vertical axis of the recorded streak images serves as the time axis whereas the horizontal one corresponds to the wavelength axis. Temporal profiles of the streak camera images were extracted by placing vertical sampling windows at the desired wavelength positions. Imaging of the laser light on the sample surface and of the pumped area was obtained using a spatially filtered pump beam, by means of a lens which projected the image onto a CCD analyzer (Newport LBP-3) with a lateral magnification value of one.

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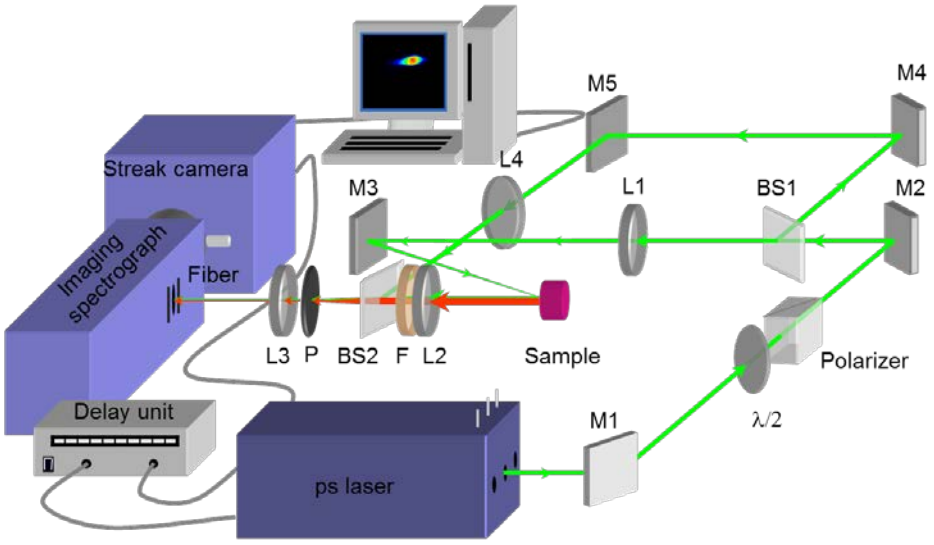

Fig. 1. Scheme of the experimental set up used to perform the ps time-resolved study of random lasing (L1, L2, L3, L4 are lenses, M1, M2, M3, M4 and M5 are mirrors, BS1 and BS2 are beam splitters, F is a long-pass filter and P is a pinhole).

3. Results and discussion

In the following, the modal behavior of a DRL is investigated not only spectrally, as most experimental works reported in the literature do, but also temporally and spatially.

3.1 Mode selection based on spatial filtering

Experimentally, the lasing modes are identified by narrow emission features in the emission spectrum. The reduction of the number of random lasing modes which are correlated in space, enables us to discriminate between different modes and therefore study their structure and dynamics. For that purpose, we control the size of the area whose emitted radiation is collected with spatial filters of different apertures. The comparison of streak images recorded by using different pinholes at a single position on the sample and at the same excitation energy above threshold, evidences the effect of the spatial filter size on the laser spike distribution. As an example, Fig. 2 shows on the left, the 3D map surfaces of the RL pulse obtained from single streak images of the explored powder, recorded at 5.5 μJ/pulse without any pinhole (a) and by using spatial filter diameters of 50 μm (b), 30 μm (c), and 15 μm (d). We should remark that in the absence of pinhole, a smaller gain of the camera was employed in order to avoid camera saturation. Additionally, under the employed focusing conditions, the onset of RL action (which is experimentally determined by the temporal shortening and spectral collapse of the emission output) is around 1.6 μJ/pulse. As can be observed in this figure, random lasing occurs in many modes which turn on at different times. Nevertheless, one can effectively reduce the number of those supported (while maintaining the same diameter of the gain medium) by decreasing the aperture of the pinhole. In fact, when the pump power is near the threshold and a pinhole of 15 μm diameter is employed, isolated spikes in the RL emission of the explored sample are observed (see Fig. 2(d)). This reveals that single modes originated from non overlapping cavities start to lase. The corresponding time-integrated spectra are presented in the XZ plane of such figures. It is important to notice that these data do not provide any insight on the dynamics of an RL system. Nevertheless, this is the kind of spectrum typically provided in almost all RL experiments reported in the literature with pulse laser experiments.

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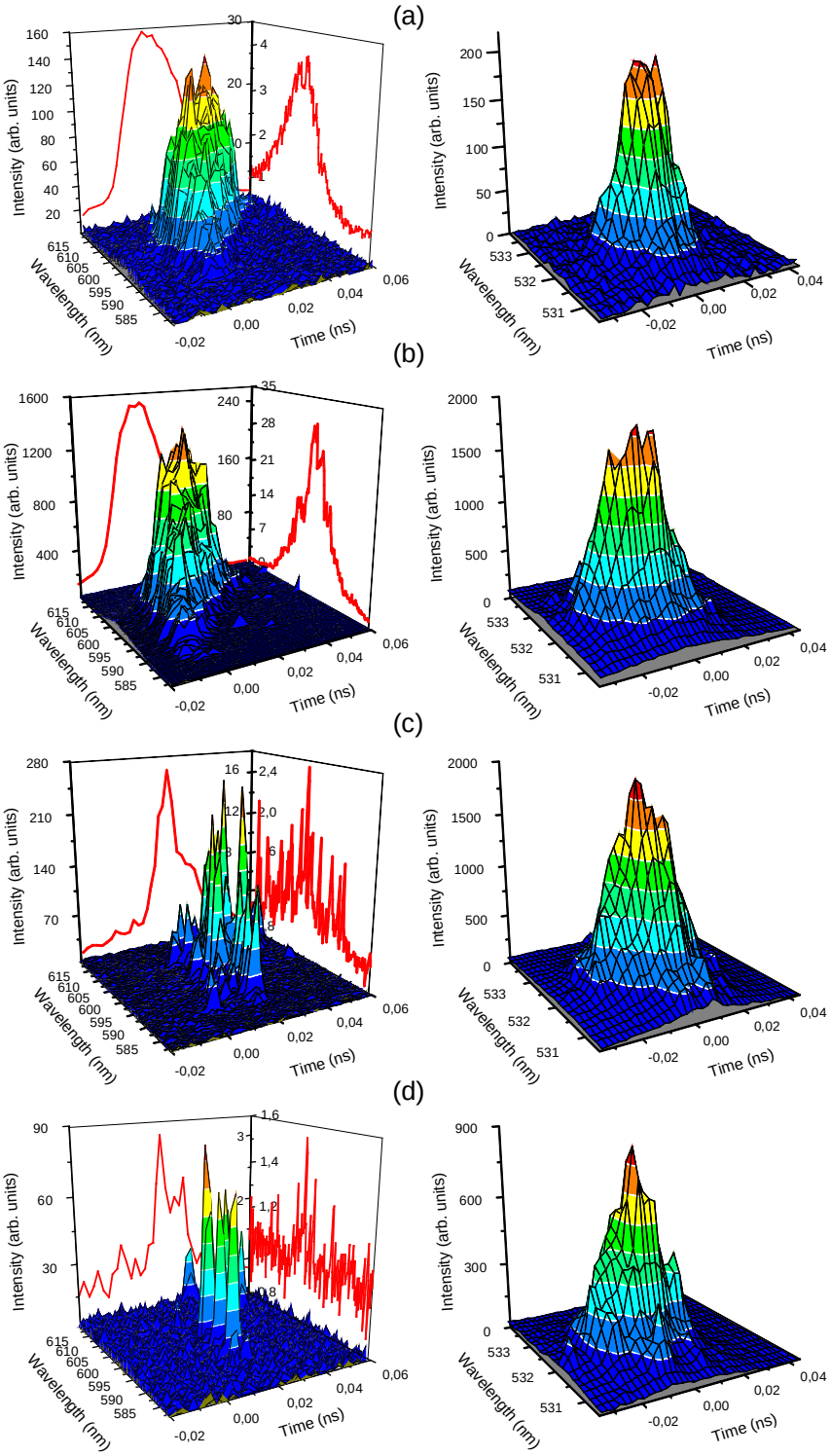

Fig. 2. 3D map surfaces of the RL (left) and pumping pulses (right) obtained without a pinhole (a) and with a $Ø_{pinhole}$ = 50 µm (b), 30 µm (c), and 15 µm (d) at 5.5 µJ/pulse. Spectral and temporal profiles extracted over the whole images are presented in the XZ and YZ planes of the left figures, respectively.

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The spectrum shown in Fig. 2(a) just presents a small fine structure on top of the ASE band. Due to the large number of spatially overlapping lasing modes existing at 5.5 μJ/pulse and detected without a pinhole, their average spectral widths exceed the average spacing between neighboring modes. As a result, spikes assigned to discrete lasing modes are averaged out leaving the smooth spectrum presented in Fig. 2(a). A spikier spectrum is found when a pinhole of a 50 μm diameter is employed (see Fig. 2(b)), whereas a clear fine structure is observed in the spectra presented in Figs. 2(c) and 2(d). The corresponding temporal profiles extracted over the whole streak images are depicted in the YZ planes of these figures. These curves become again smoother as the diameter of the spatial filter increases. Note also that Fig. 2 includes the map surface of the corresponding pumping pulses whose centers of gravity serve as time reference. The similar and smooth shape of all of them, regardless the employed pinhole aperture, confirms the stability and monomode condition of the pump source. Alternatively, a reduction of the RL spike density was achieved through laser beam focusing. If the size of the pumped area is reduced, the subset of the probed scattering medium diminishes. As a result, the number of available modes which could spatially overlap decreases, and the emission spectrum displays clear well separated sharp peaks.

3.2 Lasing features upon repeated identical excitations

We have also carefully analyzed the spectral position of spikes for different shots under identical experimental conditions. We seek to know whether the frequency of the lasing peaks is completely determined by the spatial distribution of the dielectric constant, *i.e.* by the realization of disorder, or not. In the first case, no change in the mode frequency of an RL with static disorder (like the one explored here) is expected from shot-to-shot. Sequences of several shots were recorded under the same experimental conditions while pumping at the same position on the powder sample. As an example, Fig. 3 shows the map surfaces of the RL and pumping pulses obtained with identical experimental conditions of Fig. 2 but for other shots of the same sequences. Map surfaces obtained with the same pinhole size (see Figs. 2 and 3) evidence almost no pulse-to-pulse variation of the pumping pulse while the spectral position of spikes differs individually from shot-to-shot. These fluctuations are therefore intrinsic to the random lasing and not just the manifestation of instabilities in the pump energy. In fact, when discrete modes become visible in the RL output, one can clearly observe that no specific frequency dominates the others and that spikes are randomly distributed in time and wavelength from pulse-to-pulse. The system lases in different modes in consecutive excitation events. Such a stochastic behavior which is thus not related to the type of disorder, was also reported in some previous works and explained on the grounds of strongly coupled modes triggered by spontaneous emission. In [16], time-integrated spectra of an RL with static disorder obtained under controlled and stable experimental conditions, were examined in detail. There, it was found that the location of spikes for successive emission spectra was not correlated and neither were they equispaced. The spectral profiles we extracted for comparison over the whole time range of the streak images show exactly the same behavior (see spectra presented in the XZ planes of Figs. 2 and 3). No qualitative agreement is thus found between these experimental results and theoretical predictions of the multimode lasing theory proposed by Türeci *et al.* [30, 31]. Consequently, the mentioned theoretical approach might not be suitable to rigorously treat our DRL. In contrast, other experimental studies performed on materials with static disorder, reported rather stable frequencies of the lasing peaks between different pump pulses, whereas modal intensities widely vary [33].

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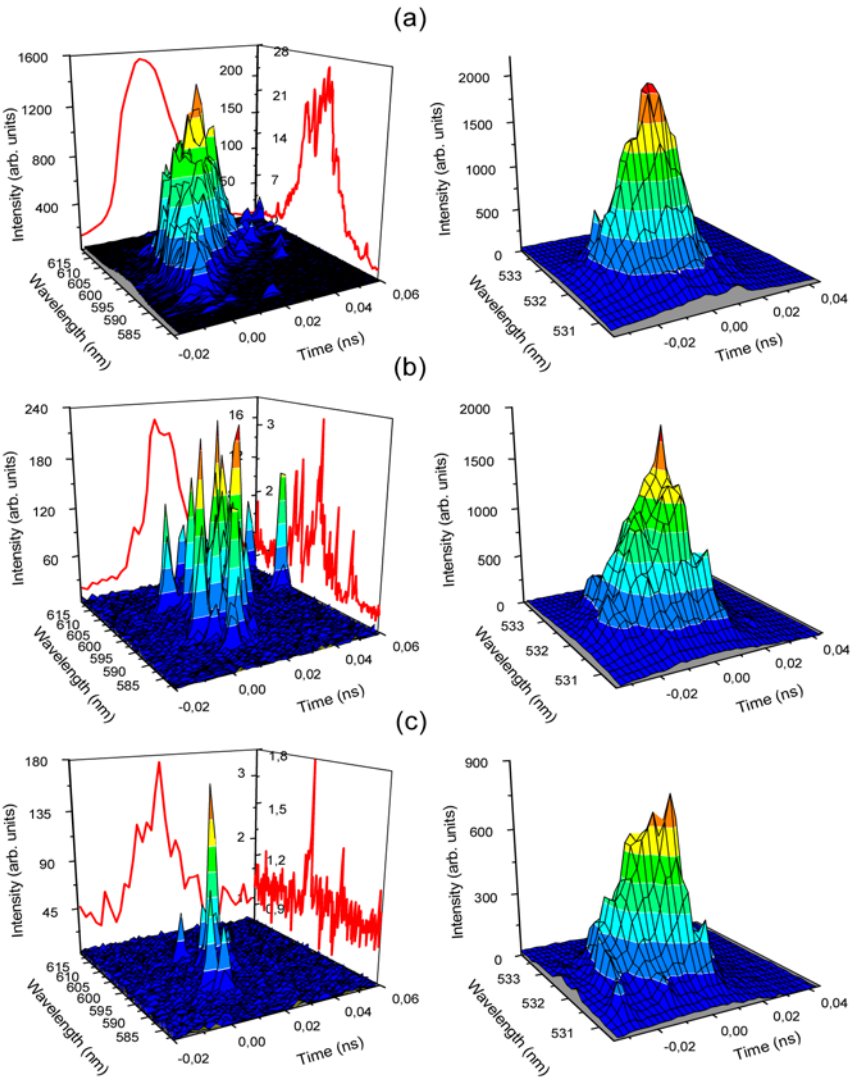

Fig. 3. 3D map surfaces of the RL (left) and pumping pulses (right) obtained with a $Ø_{pinhole}$ = 50 µm (a), 30 µm (b), and 15 µm (c) while keeping constant the experimental conditions used for Fig. 2 but in different shots. Spectral and temporal profiles extracted over the whole images are presented in the XZ and YZ planes of the left figures, respectively.

Only time-resolved measurements can explore the dependence of the observed stochastic nature of lasing spikes also on the time scale. The examination of the obtained RL map surfaces reveals different temporal behaviors of the lasing modes from shot-to-shot. Figure 4 shows some temporal profiles extracted close to the RhB gain maximum from the streak camera images whose RL map surfaces are presented in Figs. 2(c) and 3(b). For doing so, vertical sampling windows with a spectral width of 0.5 nm were employed. Note that within the spectral resolution of our spectrometer, the average spectral linewidth of the lasing spikes is 0.35 nm. This resolution was limited by the pixel size of the CCD detector (each pixel corresponded to a wavelength interval of 0.18 nm). The comparison among these time traces (obtained under an identical system configuration) shows that sharp peaks emerge at different frequencies and those which appear at the same, or at a very close, wavelength are turned on at different times. These results suggest that they are originated from different cavities.

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Moreover, the analysis of the temporal evolution of lasing spikes uncoupled from the mode competition reveals that laser modes at larger wavelengths start later whereas the average full width at half maximum of lasing modes (FWHM) is around 6 ps. Furthermore, the time traces of some uncoupled lasing modes show clear relaxation oscillations (see Fig. 4). Account taken that the average separation between adjacent peaks of each lasing mode trace gives a quantitative determination of the oscillation period, we computed from this figure different oscillation periods for different lasing modes, which again indicates they are from different cavities. Relaxation oscillation periods in the 8-38 ps range were found. It is worthy to remark that in strongly coupled modes, their originally independent relaxation oscillations can be coupled and generate new oscillations [20]. Therefore, only an efficient mode-selective approach such as the one proposed here, can provide experimental evidence of the dynamic response and relaxation oscillations of individual RL modes.

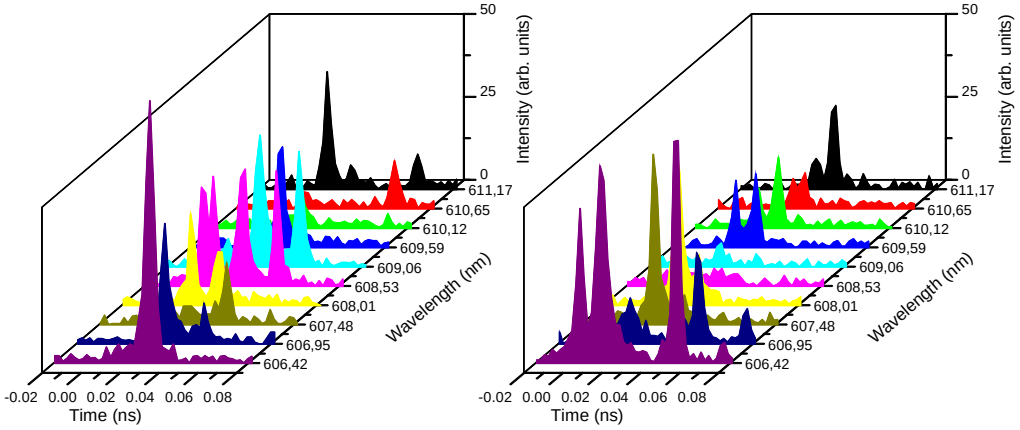

Fig. 4. Time traces of different RL modes extracted from the streak images whose 3D map surfaces are presented in Fig. 2(c) (left) and Fig. 3(b) (right). They correspond to successive excitation events with constant experimental conditions.

3.3 Effect of the pump energy on the modal distribution

The next feature we address is the pump energy dependence of the modal density in the studied powder. In Fig. 5, we look at the RL map surfaces extracted from single shot streak images recorded at 5.5 µJ/pulse (a), 27 µJ/pulse (b), and 83 µJ/pulse (c), being 30 µm the pinhole diameter. It is clear from this figure that apart from the enhancement of the spikes intensity, our system tends to emit a larger number of modes when the pump is increased. The intensification of the excitation energy allows the creation of lasing modes with larger thresholds. Therefore, an enhancement with the pumping strength of the mode coupling and of the overlap between lasing spikes which tend to average them out into a smoother spectrum, is expected. This is the exact behavior found when comparing the integrated profiles shown in Fig. 5 for the different pumping energies. At the greatest one, the appearance of RL spikes is compromised by the excessive generation of modes leading to an emission line shape which resembles the smooth emission profile of a DRL. As mentioned in some reports, pumping increase can eventually saturate the number of lasing modes, due to strong modal interactions. This effect is related to the non-monotonic dependence of mode intensities on the pump strength and the complete disappearance of some modes for pumping exceeding certain threshold. This mode suppression reported in [30], was not observed in our powder hybrid material in the employed energy range. Moreover, between 5.5 and 83 µJ/pulse and within our time resolution, no significant change of the time delay between the pumping pulse and the RL pulse (known as RL build-up time) was observed. An RL build-up time of around 12 ps was found in all cases.

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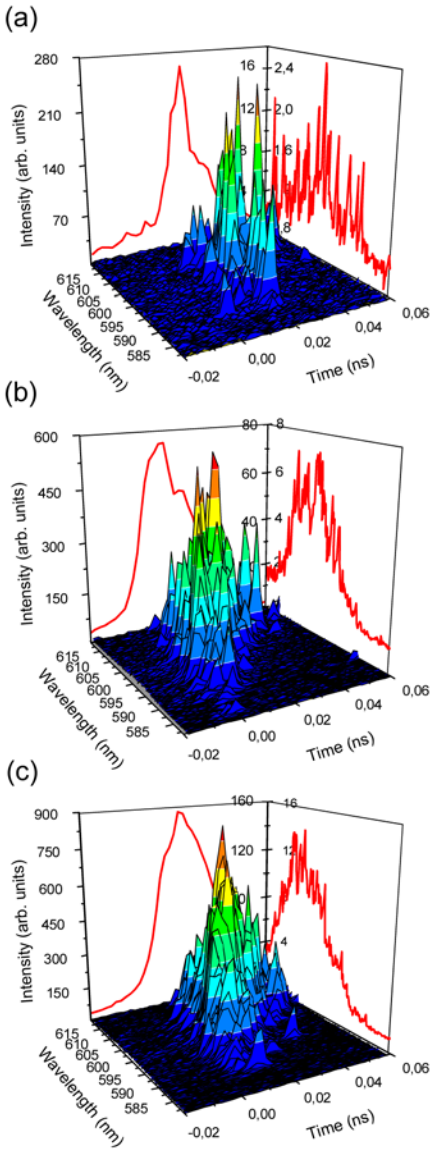

Fig. 5. 3D map surfaces of the RL pulse measured with a $Ø_{pinhole}$ = 30 µm at 5.5 µJ/pulse (a), 27 µJ/pulse (b), and 83 µJ/pulse (c). Spectral and temporal profiles extracted over the whole images are presented in the XZ and YZ planes of the figures.

3.4 Spatial features of the RL emission

Finally, we investigate the spatial features of the RL action in the explored powder sample. For that purpose, imaging of the excited region and of the emission intensity distribution on the sample surface was performed under different focusing conditions and excitation energies. As an example, Fig. 6 shows the images recorded with the CCD analyzer pumping below (Figs. 6(a) and (b)) and above (Figs. 6(c) and (d)) the RL threshold. Sizes of pumped (left) and emission (right) areas are given in the figure. These images reveal that the emitted light is extended over a larger region than the excited zone regardless the pumping energy. This result provides some sort of clue about the dominant role played by diffusion processes over amplification or spontaneous emission processes in the RhB doped hybrid powder. Identical

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results were found by focusing the laser beam to other spot sizes. Contrary to our findings, other imaging experiments of laser light on the surface of different random media revealed that lasing modes are spatially restricted to the pumped domain with an exponential tail outside it [46]. A numerical study carried out by Yamilov *et al.* showed that re-absorption of emitted light, which can be relevant in Rhodamine doped systems, suppresses feedback in the unpumped region, effectively reducing the system size [47]. Photons that are emitted in the excited region may diffuse into the surrounding unpumped volume and be absorbed there, so the probability of light returning to the pumped region diminishes. Yamilov *et al.* found in consequence, a confinement of lasing modes inside the excited domain in diffusive random media with re-absorption. In contrast, Fig. 6 clearly shows that the above mentioned spatial restriction of RL modes does not occur in our powder but instead, they are extended over a larger zone of the scattering medium.

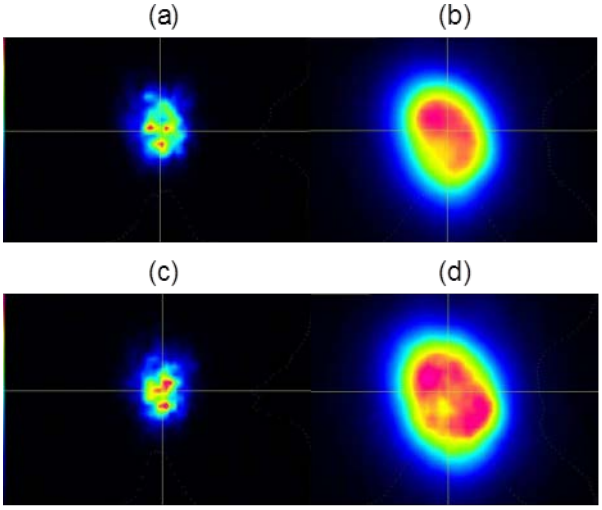

Fig. 6. Images of the emitting surface of the RhB doped di-ureasil powder pumped at 0.2 µJ/pulse (upper) and 10 µJ/pulse (down), *i.e.* below and above RL threshold, respectively. The left hand images (a, c) are images of the pumped area, since sample emission has been removed by filtering. Those to the right (b, d) are images of the emission area, since the reflected or re-emitted pumping has been removed by filtering. Widths of beams at 37% of maximal intensity are: a) 126 × 137 µm, b) 264 × 294 µm, c) 105 × 132 µm and d) 301 × 329 µm.

4. Summary and conclusions

In the present work, we develop an experimental method based on spatial filtering that allows us to study the dynamics and structure of individual lasing modes of a random laser with diffusive scattering, in particular, of a Rhodamine B doped di-ureasil hybrid ground powder. In contrast to conventional approaches, we herein perform an accurate time-resolved analysis of laser emission by the use of a streak camera. The outcome of our experiments shows that the structure of its random laser emission output is critically affected by the dimensions of the regarded emitting area or the pumping energy. These observations can be understood in terms of mode coupling. The reduction of the spatial filter aperture used to control the size of the detected emitting surface area, allows us to reduce the number of spatially overlapping random laser modes (while maintaining the size of the gain medium). We have demonstrated that this mode selection technique makes it possible to characterize the temporal and spectral characteristics of individual lasing modes as it enables the observation of discrete lasing spikes originated from non overlapping cavities. Conversely, if no pinhole is used, the number of supported lasing modes is so large that the fine structure of the emission line shape is partially washed out, resembling the smooth emission profile of a diffusive random laser.

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An additional reduction of the lasing peaks density can be achieved with a tighter focused beam. Furthermore, the results of our study illustrate the enhancement of the modal density with the intensification of the excitation energy as the creation of lasing modes with larger thresholds becomes then permitted. Note that within the pumping range used, the random laser build-up time keeps constant at 12 ps and no saturation effect leading to any mode suppression is observed. On the other hand, image sequences of several pumping shots recorded under the same experimental conditions unambiguously show an intrinsically stochastic behavior not only in the spectral but also in the temporal response of the random lasing of our sample with static disorder. The spectral position of spikes, which is thus not related to the realization of disorder, differs individually from shot-to-shot although almost no pulse-to-pulse variation of the pumping pulse occurs. This gives direct experimental evidence of how the system lases in different modes in successive excitation events. Note that our results agree with some previous experimental works where time-integrated data are recorded but are not in accordance with the behavior predicted by the time-independent constant-flux theory where uniformly spaced frequency spectra are expected. Moreover, the comparison of time traces extracted at the same spectral position reveals that lasing modes are turned on at different times in different shots, which suggests they are from different cavities. The temporal profiles of uncoupled lasing modes show that their average FWHM is around 6 ps showing clear relaxation oscillations with periods in the 8-38 ps range. This work thus provides a unique physical insight into the modal dynamics of a diffusive random laser which is hardly accessible and therefore rarely addressed in the literature. Most researchers investigate the steady-state properties for the sake of simplicity, but for a full understanding of the intricate behavior and mode competition of these disordered active media, it is essential to shed light on this fundamental issue. Finally, imaging results in this paper underline the dominance of diffusion over amplification processes on the spatial features of the powder random laser emission. The emitted light is extended over a larger area than the pump domain, so contrarily to what some authors assert, no spatial restriction of the random laser modes to the excited region is found in our case.

We hope this work will stimulate further theoretical and experimental investigations in finding methods to explore the temporal evolution and spatial features of the intriguing random laser phenomena in diffusive scattering systems with either a spiky or a smooth emission structure.

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