

## Light Disorder as a Degree of Randomness to Improve the Performance of Random Lasers

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 (Received 25 January 2021; revised 19 May 2021; accepted 8 June 2021; published 24 June 2021)

The operation of random lasers (RLs) is possible due to multiple light scattering, which provides optical feedback inside the gain medium. No conventional optical cavity is required for the operation of RLs and, therefore, RLs provide the best evidence to defy the conventional view that disorder is an unwanted phenomenon when present in physical systems. Mirrorless lasers demonstrate that the degree of disorder, associated with the spatial distribution of scattering particles, is the main factor responsible for generating a light source with low spatial coherence; this is reason why RLs are so attractive for modern imaging systems. Here, we demonstrate how to optimize the performance of typical diffusive RLs by exploiting a different degree of randomness in the system, which is loaded onto an induced disorder in the transverse intensity and wave-vector distributions of the light pattern that pumps the random-lasing medium. Quantitative analyses show that the RL's intensity emission and threshold fluence can be optimized by more than 20 times, when excited by an optical field presenting a random spatial-intensity distribution, with high speckle contrast, becoming an efficient methodology to induce high photon degeneracy in RLs required for high sensitivity applications.

DOI: [10.1103/PhysRevApplied.15.064062](https://doi.org/10.1103/PhysRevApplied.15.064062)

### I. INTRODUCTION

Disorder is a ubiquitous phenomenon, although not always desired, in optical and photonic systems. Its unpredictable behavior is the main reason why disorder has long been considered as a limiting factor for the study of light-matter interaction phenomena, as well as limiting the performance of optical and photonic devices [1,2]. In particular, light scattering is one of the most typical phenomena induced by the intrinsic disorder of photonic systems that interact with electromagnetic fields. The advantages (or disadvantages) of the scattering phenomenon depend on the type of application being explored. For instance, when light scattering is caused by dust or system imperfections, its contribution is unwanted, becoming critically detrimental for various applications, such as optical communication and high-resolution imaging systems [3,4]. However, understanding the physical properties that can be controlled through the degree of disorder in a system allows their contributions and induced phenomena to be desired and necessary [5–8].

Random lasers (RLs) are photonic systems that take most advantage of disorder. Unlike conventional lasers that use optical cavities to trap and amplify light, diffusive RLs were conceived to achieve optical feedback provided by the light scattered from disordered nano- or microstructures present inside a gain medium [9–11]. Therefore, the disordered spatial distribution of scattering particles allows RLs to emit simultaneously a large number of optical modes with uncorrelated phases. Hence, features such as moderate spectral density, low spatial coherence, and broad and complex angular emission are commonly obtained in RLs and managed by changing the active (or passive) medium and the pumping laser. In particular, the degree of disorder of the scattering gain medium is one of the most exploited parameters to control the performance of RLs. For instance, when incorporating scattering elements with different concentrations that are randomly located inside a laser medium, control over the RL wavelength, as well as over the position, number, and size of laser modes, is reported [12,13]. Also, different disorder configurations can lead to transitions between the diffusive RL regime and the Anderson-localized RLs [14], resulting in control over the coherence and directionality of the emitted light [15,16]. The polarization of RLs is also

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a parameter that can be manipulated through the degree of disorder of the scattering medium, becoming a high-value tool for the manufacture of polarization-maintaining random-fiber lasers [17].

The influence of disorder in photonic systems is not unique to the scattering medium but can also have an impact through the proper selection of the pumping light. The speckle patterns are a clear manifestation of the disorder imprinted on the intensity and phase of the optical fields, which display important contributions to interference of scattered light, optical-image processing, and displacements and deformations of diffuse objects and biological tissues [18–21]. Concerning RLs, theoretical proposals argue that their optical emission can be strongly influenced by introducing randomness into the light that pumps the lasing medium [22]. For example, active control of the spatial-intensity distribution of the pump laser can allow the selection of different random-lasing modes [23], as well as achieve highly directional emission [24]. An experimental corroboration of the efficiency of this methodology was reported in an optofluidic RL, where it was possible to obtain single-mode laser operation at different wavelengths [25].

Among the various applications proposed and studied for RLs, it is in the development of modern imaging systems that require high sensitivity and large spatial resolution where they have achieved their greatest potential [10,26]. Their main advantage over conventional lasers lies in the low spatial coherence caused by optical feedback supplied by disorder-induced scattering, which reduces the interference processes of mutually coherent photons that suffered random phase delays due to interaction with a rough surface [27]. Therefore, disorder in RLs is responsible by suppressing the formation of coherent image artifacts, such as interference-induced speckle, generating high-quality images in dimensions around the diffraction limit. However, the efficiency of a laser imaging system requires that the light source, in addition to low spatial coherence, emits many photons per coherence volume. This last condition is not always achieved in diffusive RLs, especially those that are pumped by an external laser with low intensity and a low repetition rate. Here, we show that, although disorder corresponding to the spatial distribution of scattering particles is not enough to optimize both conditions, an unusual degree of randomness in the RLs, which is introduced by disordered light patterns that pump the lasing medium, is capable of producing high photon degeneracy, strengthening their potential application in sensitive imaging systems.

## II. RESULTS AND DISCUSSION

### A. Light patterns with different degrees of disorder

In the experiments reported here, the randomness of the excitation laser is associated with its random intensity

distribution along the transverse-beam profile. This is achieved by passing a Gaussian laser beam through light-shaping diffusers, generating a speckle pattern constituted by intensity hot spots with random spatial distributions, as shown in Fig. 1(a). The size and transverse location of the hot spots depend on both the coherence of the pump laser and the scattering degree of the diffusers [28]. Four diffusers, with different diffusion angles, and designed in such a way as to present a normal distribution of the field components, are used. Images of the transverse-intensity profiles in the plane of incidence of the lasing media, captured by a CCD camera, with and without the presence of the diffusers, are displayed in Figs. 1(b)–1(f).

The degree of disorder of the light that pumps the RL is quantified by determining the speckle contrast,  $C = \sigma_I / \langle I \rangle$ , where  $\sigma_I$  and  $\langle I \rangle$  are the standard deviation and the average intensity of the speckle patterns, respectively, obtained from 100 consecutive measurements using the same diffuser. Speckle contrast can assume values ranging from  $C = 1$ , for a perfectly coherent speckle pattern, to  $C = 0$  (completely incoherent), where the speckle pattern is lost, becoming a uniform image. Statistical analysis of the images is carried out by considering the central part of the intensity profile, using an intensity matrix of  $400 \times 400$  pixels, corresponding to the smallest area containing enough scattering centers for the analysis to remain unaltered. It is important to mention that, because the properties of the pump light source, such as intensity and coherence, are kept constant during all measurements, the randomness in the spatial intensity and wave-vector distributions of the incident light is directly associated with the inherent degree of scattering of the diffusers.

A progressive increase in the speckle contrast of Figs. 1(c)–1(f) is measured as a function of the decrease in the diffusion angle ( $10^\circ$ ,  $5^\circ$ ,  $1^\circ$ ,  $0.5^\circ$ ), with contrast values of 0.15, 0.18, 0.37, and 0.55, respectively. A higher speckle contrast is interpreted as a higher degree of spatial coherence of the light patterns that pump the RL. The pumping pattern that displays a transverse Gaussian intensity distribution [Fig. 1(b)] shows a contrast of 0.08. However, because the laser beam does not pass through any diffusers, the speckle contrast loses its physical meaning and cannot be interpreted as a result close to that of a completely incoherent pattern ( $C = 0$ ). In this situation, the contrast represents exclusively a measure of the low degree of inhomogeneity of the cross-section intensity distribution, which is used as a reference to compare the influence of the degree of disorder of pump light on the performance of RLs.

For the sake of completeness, in Figs. 1(g) and 1(h), the intensity-probability density function,  $P(I)$ , and the 2D spatial-intensity correlation function,  $C_I(|\Delta r|)$ , are plotted from the transverse-intensity profiles of Figs. 1(c)–1(f). For the disordered light pattern with the highest degree of spatial coherence ( $C = 0.55$ ),  $P(I)$  behaves as a function

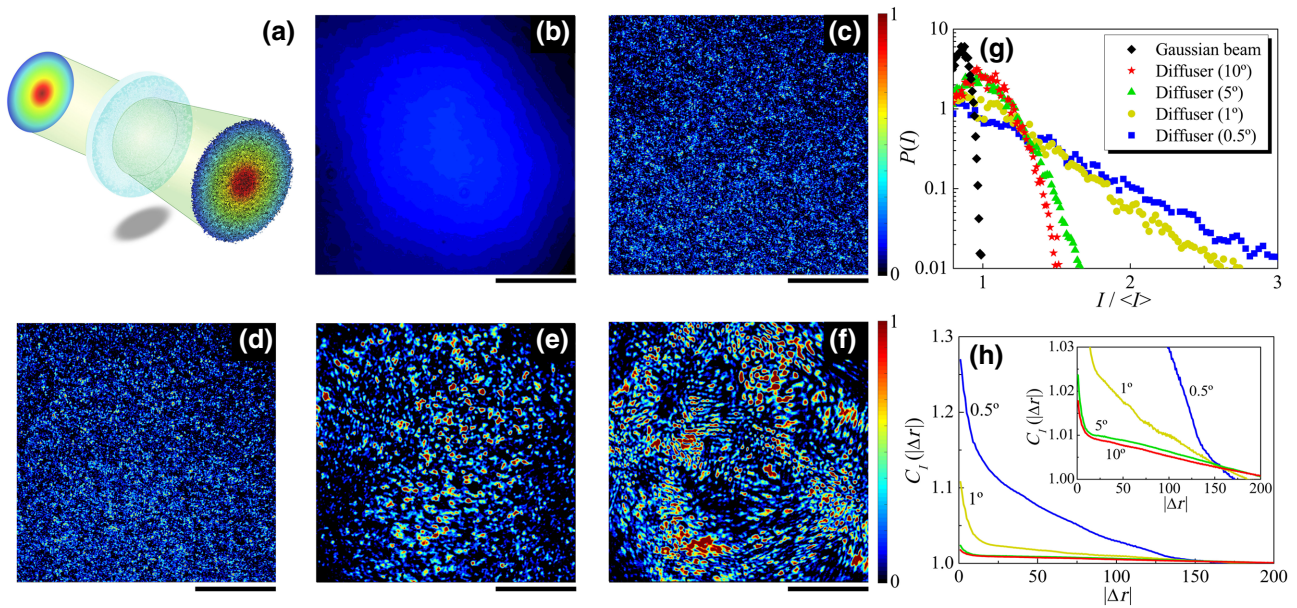


FIG. 1. (a) Experimental scheme for disordered light pattern generation, and transverse-intensity profiles, in the input face of the sample, for (b) Gaussian beam and (c)–(f) speckle patterns with diffusion angles of (c) 10°, (d) 5°, (e) 1°, and (f) 0.5° (scale bar: 1 mm). (g) Intensity probability density function, and (h) two-dimensional spatial intensity-correlation function of pump-beam profiles. Numbers on the curve represent diffusion angles of diffusers.

close to a straight line, on a semilogarithmic scale [see Fig. 1(g)], which corresponds to a partially coherent speckle pattern. The greater width of  $P(I)$  shows that light with a speckle contrast of 0.55 is constituted by a significant number of small spatial regions with high intensity ( $I/\langle I \rangle$ ). On the contrary, with increasing degree of light scattering, as induced by diffusers with higher diffusion angles, it is well known that  $P(I)$  gradually changes for a delta function [29]. This behavior can be observed through statistical analysis of the light patterns with speckle contrasts of 0.37, 0.18, and 0.15, as shown in Fig. 1(g). Light with lower intensity contrast, such as the Gaussian profile ( $C = 0.08$ ), have a more homogeneous intensity distribution concentrated in the lower values of  $I/\langle I \rangle$ . The 2D spatial intensity correlation function,  $C_I(\Delta r) = \frac{\langle \int d^2r I(r)I(r+\Delta r) \rangle}{\langle \int d^2r I(r) \rangle \langle \int d^2r I(r+\Delta r) \rangle}$ , shown in Fig. 1(h), corroborates the interpretation that disordered light patterns with higher (lower) speckle contrast display higher (lower) spatial correlation and, consequently, a higher (lower) degree of coherence, as discussed for the  $P(I)$ .

**B. Influence of disordered light on the performance of diffusive RLs**

The influence of the randomness of the pump light on the performance of the RLs is evaluated using the experimental setup shown in Fig. 2. The second harmonic of a Q-switched Nd:YAG laser (7 ns, 532 nm, 10 Hz) is used as a pump source to excite the RL medium. Control of the total power and linear polarization of the incident beam is

accomplished by means of a half-wave plate followed by a Glan-Taylor polarizer. To obtain a well-behaved Gaussian transverse-intensity profile, corresponding to a typical case of RL excitation, the laser beam is passed through a spatial filter, giving rise to a light pattern with homogeneous intensity distribution ( $C = 0.08$ ). The increase in speckle contrast, from 0.15 to 0.55, due to the disordered light-intensity distribution is managed by using four transparent light-shaping diffusers with diffusion angles of 10°, 5°, 1°, and 0.5°. To illuminate the lasing medium with a constant beam size (volume gain) in all experiments, a collimating and focusing optic, composed of two lenses with focal lengths and positions chosen to compensate the different diffusion angles of the speckle patterns, is mounted. This condition is essential to compare the behavior of

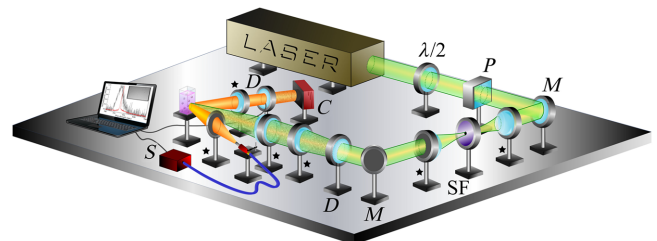


FIG. 2. Experimental setup for the study of RLs pumped with disordered light patterns, with the following optical components: polarizer (P), mirror (M), spatial filter (SF), light-shaping diffuser (D), CCD camera (C), and spectrometer (S). All components labeled with a star represent spherical lenses.

RLs against the disorder of light with a constant excitation volume of the samples, that is, without variation in the number of scattering particles that contribute to the lasing phenomenon. The Gaussian beam and disordered light patterns exhibit a total diameter of 3 mm at the sample entrance plane; the light patterns contain individual speckles with mean sizes of 22, 26, 49, and 56  $\mu\text{m}$  were measured from Figs. 1(c)–1(f), respectively. The input-beam fluence in the RL experiments is determined by dividing the energy per pulse by the total area of the light patterns.

The RL emission is collected by two detection systems, at an angle of  $45^\circ$  to the incident-beam direction. One system is used to measure the RL spectral density and spectral width through a high-resolution spectrometer (0.27 nm). Simultaneously, the other detection system analyzes the formation of speckle patterns using Köhler illumination, as described in Ref. [10]. Both the CCD and spectrometer are triggered by the flashlamp of the pump laser. Thus, spectra and images are collected pulse by pulse, together with the laser signal acquired using a reference photodetector. Post-experiment spectral and image processing are performed to select measurements with variations in pump fluence of, at most, 7%. Figure 3(a) shows the results for both detection systems when the lasing medium, constituted of 120-nm silica nanoparticle (NPs) ( $\rho = 4.4 \times 10^{12}$  particles/ml) suspended in an ethanolic solution of Rhodamine 6G (Rh 6G at 2 mM), is pumped by a disordered light-intensity distribution with the highest speckle contrast ( $C = 0.55$ ). From the point of view of characterization of the RL action, a typical transition from luminescence to the lasing regime is observed with the increase of the pump fluence, as characterized by a narrowing of the emission spectrum. For all experiments with different pump patterns, a similar redshift is observed in the RL emission peak in relation to the maximum photoluminescence, which has been previously reported in several works due to the increase in the dye and NP concentrations and pump energy (see, for example,

Ref. [30]). The evidence for a spectrum with a single peak, with a full width at half maximum (FWHM) of about 3 nm, is a signature of a RL with incoherent feedback, consistent with the condition that the scattering mean free path (approximately 450  $\mu\text{m}$ ) is less than the RL-medium thickness (10 mm). The scattering mean free path is estimated by  $l_s = 1/\rho\sigma_s$ , where  $\rho$  is the particle density and  $\sigma_s$  is the scattering cross section of a  $\text{SiO}_2$  spherical NP, obtained from the extinction spectrum. The many different spatial modes generated by this class of RL give rise to a speckle-free image [inset of Fig. 3(a)], even when the laser medium is pumped by a disordered light pattern, maintaining its low spatial coherence and high potential for laser imaging systems.

The most significant benefit of using a random intensity spatial distribution to pump diffusive RLs is revealed by the spectral RL radiance and, consequently, the RL photon degeneracy. That is, the greater the amount of radiation passing through a unit area and into a unit solid angle within a unit frequency bandwidth, the greater the RL photon degeneracy [10]. For instance, Fig. 3(b) shows that, compared with conventional excitation of RLs, using laser beams with homogeneous Gaussian intensity distribution, enhancement factors of 1.4, 1.6, 2.5, and 3.4 are obtained for spectral radiance when the transverse-intensity profile is randomly modulated to exhibit speckle contrasts of 0.15, 0.18, 0.37, and 0.55, respectively. Since the pump-beam fluence (70  $\text{mJ}/\text{cm}^2$ ) is kept constant in all experiments, we attribute the enhancement of RL performance to the disordered distribution of intensity hot spots with high speckle contrast, which concentrate a larger number of excitation photons in a relatively small volume, resulting in an increase in the number of photons emitted per coherence volume (photon degeneracy). Furthermore, Fig. 3(b) also shows that, for a pump beam with defined speckle contrast, by rotating the diffusers through the beam-propagation axis, variations of around 10% in the average RL emission intensity are produced, probably due to the emission

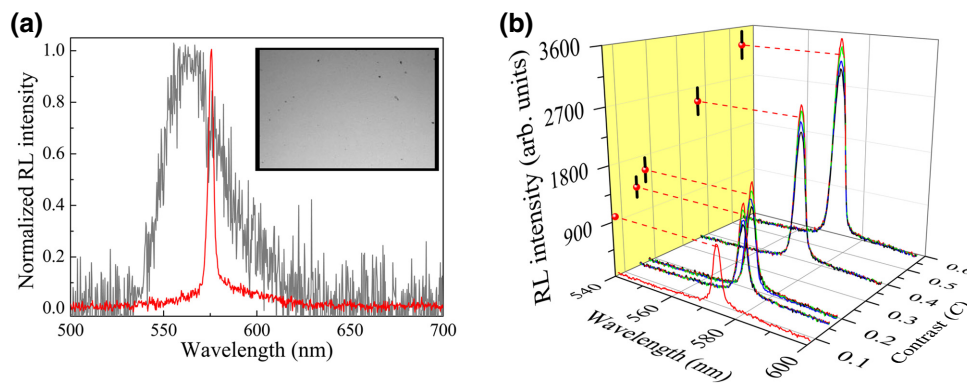


FIG. 3. (a) Normalized emission spectra below (gray line, 0.1  $\text{mJ}/\text{cm}^2$ ) and above (red line, 70  $\text{mJ}/\text{cm}^2$ ) the RL threshold, pumped by a disordered light pattern with  $C = 0.55$ . Inset: speckle-free image obtained from RL emission. (b) RL intensity pumped by different pump-beam patterns.

of new and more efficient RL modes. Therefore, the results indicate that a maximum improvement in the diffusive RL performance can originate from an ideal match between the intensity and wave-vector disorder degrees, as monitored by the speckle contrast of the pump patterns and the spatial distribution of the scattering NPs. Unfortunately, it is not possible to analyze the RL mode structures because in the incoherent feedback regime the RL emission spectrum is constituted by the superposition of a large number of spectrally and spatially overlapping modes.

The performance of the RLs pumped with disordered light patterns exhibiting different speckle contrasts is evaluated from the dependence of the emitted light intensity and the FWHM of the emission line shape as a function of the excitation-beam fluence, as shown in Fig. 4. In all cases, the spectral narrowing concomitant with the superlinear growth of spectral radiance defines a threshold pump fluence necessary to produce RL action, which varies, following linear behavior, with the contrast [Fig. 4(c)] for values between 0.08 and 0.55. For an incident fluence of 70 mJ/cm<sup>2</sup>, a maximum decrease (increase) in threshold fluence (RL intensity) around 22.9 (3.4) times, compared with pumping with Gaussian intensity distribution, is observed for  $C=0.55$ . Notice from Figs. 4(a) and 4(b) that, by pumping the RL medium with a pure Gaussian beam, even when the pump fluence increases by about 40% (reaching 100 mJ/cm<sup>2</sup>), the RL intensity is low relative to pumping with disordered light patterns, while the minimum FWHM at high fluence remains roughly constant.

Optimization of the performance of RLs is closely related to the strength of the scattering phenomenon because its optical feedback is achieved by multiple light scattering. This occurs because the photon residence time is elongated by multiple scattering, which results in light amplification during its transport, when an optical gain is introduced into a random scattering medium. Typically, the

scattering strength of a RL is controlled by manipulating the characteristics of the scattering medium, such as particle size and concentration. However, previous studies on RLs also show that the photon residence time depends on the explored gain volume along a photon’s propagation direction, causing a variation in the number of excited RL modes and the threshold energy density [31]. In our experiments, under the same conditions of the scattering medium (concentration of silica NPs), the scattering strength can be controlled by managing the initial conditions of photon propagation from the distribution of the wave vectors,  $\vec{k}$ , in the light patterns that illuminate the random-lasing medium. In principle, unlike the traditional RL pumping condition, the size of the individual speckles containing pump photons in the disordered light patterns is much smaller than the scattering mean free path. Despite the presence of several speckles, their small sizes should be unfavorable for RL excitation, since photons must experience an insufficient number of scattering events necessary to achieve optical feedback. Nevertheless, it is important to highlight that all disordered light patterns show a large  $\vec{k}$  distribution compared with a laser beam with a Gaussian intensity distribution, where the wave vectors at the waist plane are distributed in approximately the same direction. From the experimental results, we understand that the increase in the random  $\vec{k}$  distribution favors the coupling of pump photons with various modes induced by disorder in the medium, in addition to increasing the photon residence time inside the gain medium, resulting in more efficient diffusive RLs when compared with traditional systems pumped with a homogeneous light beam, as shown in Fig. 4. However, random  $\vec{k}$  distributions, such as those exhibited by speckle patterns generated from diffusers with a high degree of scattering (i.e., speckle contrast of 0.15), can affect the light-amplification conditions necessary for the gain to overcome all losses. In this sense, a pumping beam with disorder in the wave-vector distribution, but

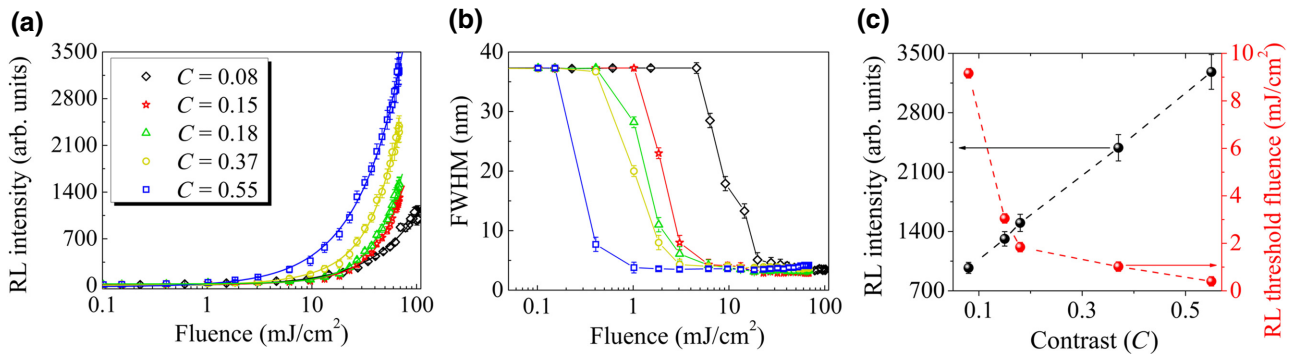


FIG. 4. (a) Emission intensity and (b) FWHM as a function of excitation fluence for RL represented by silica NPs suspended in an ethanolic solution of Rh 6G, pumped by light patterns with different speckle contrasts:  $C=0.08$  (rhombuses),  $C=0.15$  (stars),  $C=0.18$  (triangles),  $C=0.37$  (circles), and  $C=0.55$  (squares). (c) Influence of speckle contrast on RL intensity and RL threshold fluence, for a pumping energy of 70 mJ/cm<sup>2</sup>. Error bars are calculated from the analysis of 100 consecutive measurements, under the same conditions.

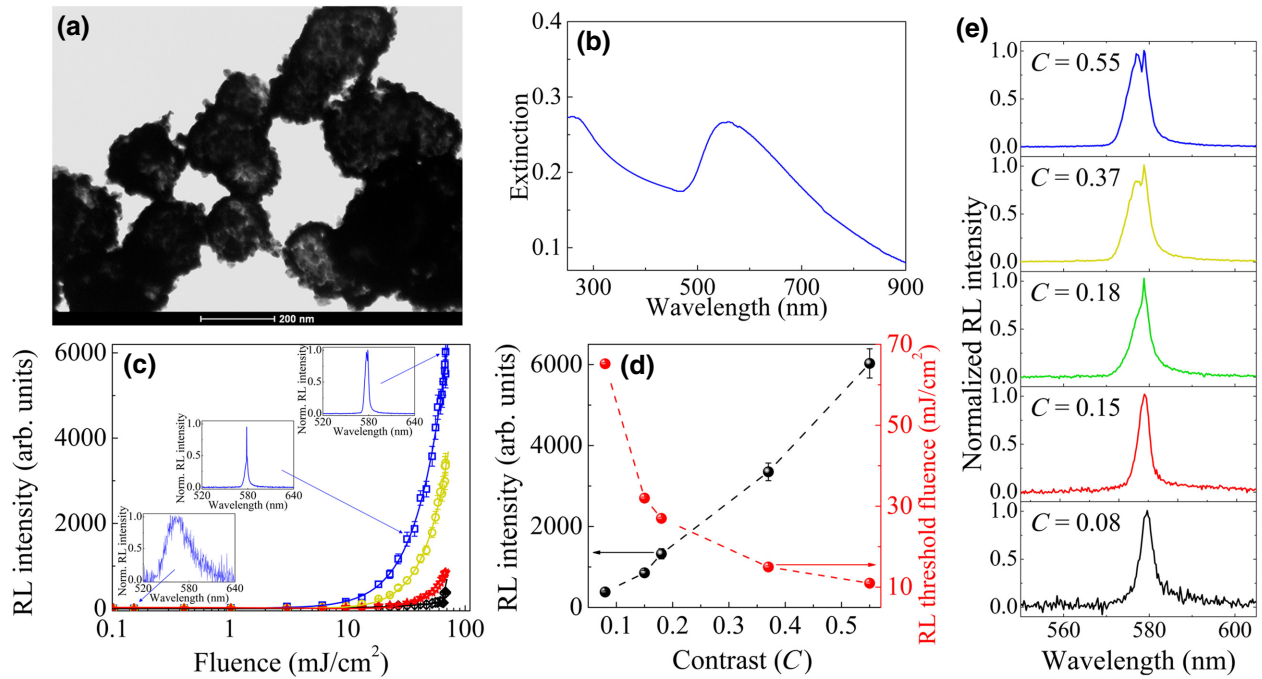


FIG. 5. (a) TEM image and (b) extinction spectrum of Si@Au nanoshells. (c) RL emission intensity as a function of excitation fluence, using Si@Au nanoshells in Rh 6G, pumped by light beams with contrasts of 0.08 (rhombuses), 0.15 (stars), 0.37 (circles), and 0.55 (squares). Insets correspond to RL emission spectra for different input fluences when pumping is performed by the beam with speckle contrast of 0.55. (d) Influence of pumping-beam contrast on RL intensity and RL threshold fluence, as well as (e) on RL intensity emission. Error bars in (c),(d) are calculated from analysis of 100 consecutive measurements, under the same conditions.

with a high speckle contrast ( $C=0.55$ ), is revealed to be the best condition to excite many laser modes with high photon degeneracy, resulting in an increase in the RL emission intensity, keeping the spectral FWHM approximately constant, and a decrease in the fluence threshold. Therefore, disorder introduced into the excitation laser beam shows clear evidence for optimization of the RL's efficiency because a lower pumping-energy supply is necessary to generate a higher RL intensity emission.

### C. Disordered light applied to plasmonic RLs

To guarantee the effectiveness of the use of this methodology, mainly in the application of RLs for imaging systems, disordered light patterns are also used to pump a more efficient lasing medium, which is obtained by attaching plasmonic NPs to the surface of the silica NPs [32], resulting in porous silica core-gold (Si@Au) nanoshell system, as shown in Fig. 5(a). In this system, fabricated as described in the Supplemental Material [33], the plasmonic effect of the gold NPs gives rise to a localized surface-plasmon band, superimposed on the extinction spectrum of the silica nanoparticles [Fig. 5(b)], which makes it possible to intensify the local field effects around the scattering NPs responsible for providing optical feedback of the RL.

Figures 5(c) and 5(d) show results like those of Fig. 4 when Si@Au nanoshells, with a concentration of

$8.8 \times 10^{10}$  particles/ml suspended in an ethanolic solution of Rhodamine 6G (2 mM), are pumped by laser pulses with different degrees of disorder on the transverse-intensity profiles. Notably, a maximum sixfold decrease in threshold fluence is obtained when the pump-laser pulses present a speckle contrast of 0.55. Such a low enhancement factor, compared with the reduction in the threshold energy by 22.9 times when only silica NPs are used as scattering particles, is understandable because the medium containing Si@Au nanoshells is 50 times more dilute to preserve colloid stability. However, under the same conditions, the simultaneous action of disorder in the pumping-light-intensity pattern and the plasmonic effect of the metal NPs leads to a 15.7-fold increase in the RL intensity emission, even exhibiting higher RL intensities than those measured with pure silica NPs.

### III. CONCLUSION

The experiments reported show that the degree of disorder in the spatial intensity and wave-vector distributions of the pump light is a powerful way to optimize the performance of diffusive RLs. Maximum optimization is found by introducing disorder into the wave-vector distribution of the pump patterns, but with a low diffusion degree sufficient to maintain a high density of pumping photons per speckle area. Both conditions guarantee better coupling of

the pump photons with different modes defined by the scattering sample and efficient pump fluence to obtain high amplification in each mode where the optical gain exceeds all losses. In a proof-of-principle experiment, a reduction of the excitation-energy threshold of a RL by more than 20 times is measured in a typical diffusion system composed of silica (scattering) NPs suspended in a laser dye solution (gain medium). Simultaneously, the RL emission intensity exhibits an enhancement of 3.4 times and is able to reach values greater than 20 times when, in addition to the speckle contrast, the plasmonic effect of metal (scattering) NPs is explored. The present work aims to introduce an unusual methodology to optimize the efficiency of various types of diffusive RLs, in which different configurations of disordered light patterns can be designed to explore low spatial coherence, to eliminate speckle, while maintaining high photon degeneracy required for high-sensitivity imaging applications.

As an extension of this work, we anticipate that the use of disordered light as a degree of randomness introduced into RLs also allows the spatial coherence degree in scattering random media based on plasmonic NPs to be managed. As observed in Fig. 5(e), unlike an excitation beam with a Gaussian transverse-intensity profile ( $C=0.08$ ), for pump beams with higher speckle contrast, it is possible to observe the appearance of two small and narrow peaks in the emission spectrum on top of a global narrowing, indicating the beginning of a transition to the coherent feedback regime [34]. Work aimed at studying the influence of various disordered light patterns to pump coherent plasmonic random lasers is currently in progress. One promising point for studies in this direction may be the development of theoretical or numerical models that analyze the influence of disordered light patterns on the RL operating mechanisms, as well as experimental studies to analyze the mode structures, polarization, and directivity of the RL emission on diffusive and localized regimes.

### ACKNOWLEDGMENTS

This work is supported by the Brazilian agencies Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Fundação de Amparo à Ciência e Tecnologia do Estado de Pernambuco (FACEPE), and Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES).

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