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PARTIALLY PUMPED RANDOM LASERS

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Conventional lasers consist of two components: a gain material that is pumped in order to provide amplification of light and a cavity to provide feedback. Random lasers replace the traditional laser cavity with a random, multiple-scattering medium. This can give rise to complex lasing behavior, such as unpredictable multidirectional and multifrequency output. Controlling these systems has proved difficult and, until now, has consisted of material and structural manipulations. In random lasers, the most common pumping mechanism is an optical field, which can be applied uniformly or partially across the scattering medium. Partial pumping, referring to the restricted spatial extent of the pump applied to the gain material, is therefore quite ubiquitous in such systems. In contrast to conventional lasers, however, the impact of partial pumping can be significant in random lasers as a subset of the scattering medium is probed. In this review, we discuss state-of-the-art investigations of partially pumped random lasers. Numerical and experimental investigations of how even a simple spot profile of the pump can dramatically alter random laser output are presented. First, the simple case of partial pumping in strongly scattering systems where laser modes are spatially confined is described. Then the most common but more difficult case of weakly scattering random lasers is considered. Here, modes are spatially extended, forcing greater mode interaction and making the random laser output more difficult to predict. Finally, we review recent works that show how the pumping degree of freedom allows a general increase in control over random lasers.

Keywords: Random lasers; laser pumping; multiple scattering.

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1. Introduction

Conventional lasers consist of an amplifying material in a resonant cavity.[1] Random lasers, meanwhile, are comprised of an amplifying material in a scattering medium. A comprehensive definition of a random laser is given[2] as

> an optical structure or material that satisfies the following two criteria: (I) light is multiply scattered owing to randomness and amplified by stimulated emission, and (II) there exists a threshold, due to the multiple scattering, above which total gain is larger than total loss.

Lasers are typically built to minimize scattering, which avoids loss out of the cavity resonance. Loss results in the requirement of a greater pump energy to reach the lasing threshold and, therefore, a less efficient laser. Scattered light is not necessarily lost from the cavity, but from that particular lasing mode. However, if scattered light can be contained inside the laser cavity long enough, light intensity of the new mode formed by scattering will be amplified by stimulated emission. This recirculation of light through scattering allows lasing in a random medium if that feedback overcomes the loss of the system. This overall system loss, however, still has a strong effect on these newly formed random laser modes. The impact of such leakage on the modes themselves and on random laser output has recently been reviewed.[3]

The focus of this paper is partially pumped random lasers. The same system leakage as that discussed above is present. Now, however, the random systems have a gain distribution that is not pumped at every spatial location where pumping is possible. For example, by focusing the pump light on a small subsection of a random laser so that the atomic gain medium within the illuminated spot is pumped and that outside is not, one realizes a partially pumped random laser. This may appear to be a narrow subset of such systems. However, considering that most random lasers are pumped optically, and that the pump light cannot typically reach all regions of a random sample uniformly, partially pumped random lasers are the standard situation, though they are not always referred to as such in the literature. Conclusions reached in those prior works that neglect theoretical considerations of partial pumping often remain valid. With pumping conditions kept constant, properties of random lasers, i.e., emission frequencies, thresholds, and output directions, can be studied independently. We shall discuss in detail, however, the intimate dependence of such properties on the pump frequency, intensity, and spatial distribution. As a simple example, a reduced pump area can still allow the formation of a random laser but the lasing thresholds increase, due to a smaller amount of gain. More specifically, the lasing threshold increase experienced by each individual mode may be different due to different spatial overlap with the gain. Therefore, understanding the part played by partial pumping allows one to predict the behavior of random lasers and exercise greater control over these devices that have largely been untamed.

In more conventional lasers, partial pumping has already been utilized for control. For example, in microcavities it can control the output directionality[4–6] and select modes,[7] including those which are electrically pumped.[8–10] In slab lasers, partial pumping has been shown to improve efficiency[11] and beam quality.[12] Microchip lasers employing toroid-shaped partial pumping can allow high pump intensity with low pump power and generate more complex modes, broadening the lasers' applicability.[13]

In this paper, we shall review some of the manipulations that have been achieved by utilizing partial pumping in random lasers. Most of the work here focuses on spot pumping. Even this simple pump profile can dramatically alter the character of lasing modes. A notable example is given where only a small region of a diffusive random system is pumped. The effective system size is reduced when absorption outside the pumped domain is significant. This can generate tightly confined lasing modes, even in such weakly scattering samples. More complex shapes of the incident pump field will also be explored, due in large part to the freedom offered by spatial light modulators. On-demand control is possible through optimization of the pump profile designed to give weight to a particular lasing mode. Even with significant spatial mode overlap, one mode can be selected for lasing, thereby dictating the random laser emission frequency.

To begin with, we shall summarize in Sec. 2 some aspects of our knowledge of random lasers that are necessary to understand the consequences of partial pumping. We focus on the different regimes of scattering and clarify the properties of the pump in random lasers. Next, we examine the common spot pumping situation. Section 3 presents consequences of spot pumping in the strongly scattering regime, where light localization can take place. The more difficult regime to study theoretically, the weakly scattering regime, is discussed in Sec. 4. Here, we explore the spatial and spectral properties of the light field within the random laser and the light emission. The many strong adjustments realizable through manipulation of the pump lead to a study of random laser control in Sec. 5. Recent theoretical and experimental work is presented that illustrates the great, but as yet largely unexplored, potential of partial pumping as a knob for tuning random lasers. Overall impressions and conclusions can be found in Sec. 6.

2. Random Lasers: Theory and Experiments

The definition of a random laser in Sec. 1 is broad enough to include many types of multiple scattering, from weak to strong. In particular, it was found that the lasing properties depend heavily on the natural resonances of the random system.[3] Lasing modes formed by scattering can be based on localized modes of the random system in the strong scattering regime or on extended modes in the weakly scattering regime. These different situations result in different behavior when pumping only part of the spatial region of the random system. Thus, we begin here with a discussion of different regimes of scattering.

All cases here consider a convexly shaped collection of scatterers, so that any light emitted out the surface of the collection is not recaptured. Moreover, the boundary is open,[14] meaning once light reaches it, leakage can occur. The location of the boundary is simply where free-space begins and scatterers are absent; no containing or guiding structure[15,16] is considered. Thus, properties of random lasers presented here primarily depend on the medium within the scatterer collection.

2.1. *Multiple scattering and optical gain*

Consider light that is scattered by optical inhomogeneities as it travels through an open random medium. Scattering may be strong and the material opaque, or it may be weak and the material transparent. No restriction is placed on material opacity in the definition of a random laser, only that scattering must take place. During propagation, light may interact with an amplifying, or gain, material. If the light is sufficiently amplified before it exits the system, in such a way that gain overcomes loss, lasing action takes place. Due to the widely varying regimes of scattering, different descriptions of the lasing process exist. Here, we begin with a brief overview of single and multiple scattering. We then outline the conditions necessary to observe random lasing in a diffusive system, which is historically the first random laser theorized and observed. It is related (though distinct) to amplified spontaneous emission (ASE), since it leads to spectral narrowing of the spontaneous emission band and has been called lasing with intensity or nonresonant feedback. We shall briefly discuss the distinctions between ASE and this “incoherent” random lasing. Next, we discuss the localization of light in more strongly scattering media, which corresponds to the existence of localized eigenstates inside the random sample. Such states can play the same part as the eigenmodes of a Fabry–Pérot cavity in a conventional laser and was initially invoked to explain the sharp laser peaks that are observed on top of the spontaneous emission band. Lasing in this case has been called lasing with amplitude or resonant feedback. However, most experimental observations of narrow laser peaks were performed in systems too weakly scattering to support localized modes. This recently lead theoreticians to realize the close correspondence between the lasing modes and the extended modes (or quasi-eigenstates) of such systems, despite the strong leakage through their open boundaries.

2.1.1. *Single and multiple scattering*

Consider a plane wave with intensity I_0 incident on a random medium. When propagating inside the sample, light is continuously scattered out of the main beam. Hence, the intensity of the incident beam decreases as a function of depth inside the sample. This is the celebrated Beer's law, which reads

$$I = I_0 \exp\left(-\frac{z}{\ell_s}\right), \tag{1}$$

where ℓ_s is the scattering mean free path, which may be thought of as the average distance between two successive scattering events. In a dilute system of scattering particles, the scattering mean free path is given by

$$\ell_s = \frac{1}{(\rho\sigma)}, \tag{2}$$

where σ is the scattering cross-section and ρ the density of scatterers. More generally, ℓ_s is dependent on the density of states and the spatial correlation $\langle\epsilon(r)\epsilon(r')\rangle \neq 0$ of the dielectric function $\epsilon(r)$, where r is the position variable.[17] Depending on the scattering differential cross-section, it can take from one to several scattering events and some associated time τ_t before the memory of the incident direction beforehand is lost. The length scale corresponding to τ_t is calculated as $\ell_t = v_g\tau_t$, where v_g is the group velocity, and is known as the transport mean free path. It can be shown that $\ell_t = \ell_s/(1 - \langle\cos\theta\rangle)$, where $\langle\cos\theta\rangle$ is the average cosine of the scattering angle. The transport mean free path may be thought of as the average distance a wave travels before its direction is completely randomized. Due to the term $\langle\cos\theta\rangle$, $\ell_t \geq \ell_s$.

The mean free paths enable us to characterize different regimes of propagation in random systems. If the size of the 3D system L is smaller than the transport mean free path ($L \lesssim \ell_t$), the system is in the ballistic regime. In this case, scattering still takes place due to randomness, but light travels out of the system before its direction can be completely randomized. If $\lambda < \ell_t < L$, the system is in the diffusive regime. In this case, light follows a random walk through the system characterized by the diffusion coefficient $D = (1/3)v\ell_t$, where v is the transport velocity. The average distance traveled before exiting is $\ell_{\text{ex}} = vL^2/D$. A third regime exists in which light may be spatially localized, where $\ell_t \sim \lambda/2\pi$, as suggested early on by John[18] and Anderson.[19] In 1D and 2D, localization is always a possibility if the system is large enough.

2.1.2. *Diffusive regime*

Historically, investigations into random lasers began in the diffusive regime. Here, the random walk of photons increases the path length before leaving the gain medium, thereby enhancing amplification. Diffusion of light in the presence of gain was first proposed by Lethokov in 1968. He introduced the energy density of photons $W(\mathbf{r}, t)$ and solved the diffusion equation[20]

$$\frac{\partial W(\mathbf{r}, t)}{\partial t} = D\nabla^2 W(\mathbf{r}, t) + \frac{vW(\mathbf{r}, t)}{\ell_g}, \tag{3}$$

where $vW(\mathbf{r}, t)/\ell_g$ is the gain term. He found a critical volume $V = L_c^3$, above which the photon energy density increases exponentially,

$$L_c^3 = \left(\frac{\ell_t \ell_g}{3}\right)^{3/2}, \tag{4}$$

where ℓ_g is the average distance a wave travels before its intensity has increased by a factor e. ℓ_g is the counterpart of the absorption length ℓ_a in a dissipative system. The result of Eq. (4) can be qualitatively understood[21] by stating that a photon must travel through the random medium long enough to generate at least one extra photon before exiting the system or, equivalently, $\ell_{\mathrm{ex}} \geq \ell_g$. Using the previously given definition of ℓ_{ex} in this inequality yields the expression (4) of L_c.

Since more amplification is provided at light frequencies closest to the atomic resonance frequency, the gain at these frequencies is first to match the loss as the pumping rate increases. Outside this frequency region, amplification is still less than the loss rate. With maximum amplification close to the maximum of the gain curve, the spectral width of the emission peak narrows. A similar phenomenon to this is ASE, where the emission spectrum is found to become narrower as the pumping rate is increased.[2,22–24] Different from ASE, however, incoherent random lasing exhibits a threshold, above which the width of the emission spectrum reduces suddenly. Meanwhile, the ASE spectrum narrows gradually and does not show such a sudden drop above a pump threshold. Moreover, ASE can take place in a system without a cavity,[25] since this is only amplification of light along a long path without feedback.[1] The diffusion model including gain and ASE can describe the narrowing of broad emission spectra, but cannot give rise to laser oscillation, since it ignores the wave nature of light and treats it as a collection of classical particles. Hence, this model cannot explain the appearance of discrete lasing peaks, which were observed in the late 1990s.[22,26]

2.1.3. *Localized regime and states of random media*

Lasing oscillation in a random medium is, in fact, possible due to the coherent feedback induced by elastic scattering of light. Since scattering is elastic, interference effects persist, even in the diffusive regime. The main assumption behind the diffusion approximation is that the path the light travels through the scattering medium is so complex that constructive and destructive interference effects are scrambled and eventually average to zero. This assumption of neglecting interference effects is sufficient in many standard situations, especially when scattering is not large, as in dilute media. However, in the opposite case of strong scattering for a fixed, finite system, such an assumption can fail with the possible occurrence of Anderson localization. In the regime of localization, spatially localized resonances with small leakage rates may serve as a basis for resonant feedback in random lasers. These modes are equivalent to those of a Fabry–Pérot cavity. Such random lasers can exhibit good coherence, but since the geometry of such localized resonances is complex, they lose the beam directivity of conventional lasers.

Before examining the weakly scattering regime in the next section, it is useful to clarify the meaning of the following terms: modes, quasimodes, quasi-bound states (QB states) and resonances, all of which are used throughout this paper. First, since random lasers are open systems, the corresponding passive systems without gain are not Hermitian. Even in the localized regime, a small amount of leakage occurs. Hence, random lasers cannot be described in terms of the eigenmodes of a Hamiltonian like the modes of a closed cavity. Instead, they are described in terms of quasimodes, QB states or resonances. The three terms are used by different authors in different contexts but, in fact, they are identical. They are eigenvectors of the S-matrix of the cavity without gain and have eigenvalues of infinity. The eigenvalues of the corresponding effective Hamiltonian are complex and the QB states grow exponentially outside the system. In other words, they are solutions of the wave equation with outgoing waves only.

Let us now consider the same system with gain, i.e., the random laser. As shown recently by Türeci *et al.*, a lasing mode with a real frequency outside the system is better expanded in the so-called "constant-flux" basis.[27–31] The constant-flux states do not diverge outside the system. Therefore, a lasing mode is distinct from a QB state of the passive cavity, even at the lasing threshold where the comparison is easier due to the absence of saturation and competition effects. The difference inside the system between a lasing mode and a single QB state is large in the weak scattering limit, decreases in the diffusive regime, and becomes negligible in the localized regime.[3] Thus, there is a close correspondence between a lasing mode and a QB state as long as one does not consider the very weak scattering limit. In particular, considering the lasing modes as a superposition of resonances is an approximation that assumes the uniformly pumped lasing modes have a close correspondence with QB states of the passive system.

2.1.4. *Weakly scattering regime*

The conditions for the realization of localization of optical waves are known to be very difficult to achieve, especially in three-dimensional systems. Careful examination of some random lasers exhibiting discrete lines that are characteristic of resonant feedback has shown that such samples were far from being in the localization regime. Hence, the presence of localized states could not be given as a reason to explain the experimental observations. Some authors have put forward that in the diffusive regime, far from localization, good resonances that manifest themselves like ring waveguides may have some nonnegligible probability to exist.[32,33] However, this probability is sensitive to the correlation of the disorder and will not exist in all weakly scattering cases (e.g., in the ballistic system studied[34] by Wu *et al.*). In other cases, how this probability compares precisely to that of finding more typical, spatially extended resonances (or superposition of resonances) that can serve as a basis for a lasing mode is unknown.

Other authors have suggested that among spontaneously emitted photons of a random system with gain, a few of them could be sufficiently "lucky" to follow a very long path before leaving the system and accumulate enough amplification to lead to random spikes in the spectrum as observed in some experiments.[35,36] However, as addressed above, amplification of noise lacks the possibility of laser oscillation and phase-coherent output. Thus, these mechanisms could not explain all observations of random lasing with resonant feedback.

As discussed above, it was realized that not only can localized states in strongly scattering media give rise to sharp laser lines, i.e., lasing with resonant feedback, but that leaky resonances in diffusive and even ballistic systems can as well. Theoretical[27–31] and numerical[34,37] studies have been devoted to the correspondence between lasing modes and the resonances. The threshold lasing mode, i.e., the first mode to lase at threshold, has been found to be almost identical to a quasi-bound state of the passive system.[3] The correspondence is excellent when losses through the boundaries are small, as in the localized regime.[38–40] The correspondence deteriorates slowly when losses increase, for instance, when reducing scattering or considering higher-threshold (generally, leakier) lasing modes. However, even with significant loss, the correspondence is still good.[34,37] The multimode regime is more complex since saturation and competition effects take place. Hence, the correspondence also deteriorates as the number of lasing modes increases.[30,31] Instead, the lasing modes become a superposition of multiple resonances.[29] As shall be discussed in Sec. 4.5, partial pumping has a similar effect.

The lesson to be taken out of these recent studies is that even in very open and lossy random lasers, the "bad" resonances, or quasi-bound states, of the corresponding passive system play a role as important as the "good" modes of the cavity of a conventional laser. Considering the complexity of such quasi-bound states,[41] it is difficult *a priori* to anticipate the action of partial pumping on the random laser modes.

2.2. *Uniform and nonuniform gain versus uniform and partial pumping*

Before addressing partial pumping, it is important to clarify the meaning of uniform pumping for a random laser. The situation with gain being located uniformly in the random medium, i.e., in both the scatterers and air gaps is easier to consider theoretically, because it avoids additional light scattering caused by the spatial inhomogeneity of gain. However, this situation is more difficult to realize experimentally. In real random lasers, pumping takes place in the active elements, which usually do not fill up the total volume, such as particles suspended in laser dye where there is no gain in the particles. This situation contrasts strongly with that of conventional lasers, where pumping is usually distributed uniformly in the gain domain.

In Fig. 1, the effects of the type of gain and pumping mentioned above are illustrated in a one-dimensional random laser. Layer thicknesses vary randomly from 10 nm to 380 nm, while the free-space wavelength of the two lasing modes is 447 nm and 443 nm, respectively. The refractive index is complex, thereby providing scattering and optical gain where specified. Solutions are found at the lasing threshold for each mode individually. This means the gain (imaginary part of the refractive index) is adjusted until it precisely compensates loss; more gain is required for leakier modes and when there are fewer active elements.

In the first case (red curves) in Fig. 1, gain is placed everywhere, in the air gaps and the scatterers, uniformly. In the second case (black curves), gain is only placed in the air gaps, so that only the air gaps can be pumped. Although gain is not placed everywhere, it is homogeneously distributed across the system, i.e., it exists in every air gap within the collection of scatterers. For this reason, the nonuniform gain case is often referred to as uniform pumping. For both the uniform and nonuniform gain cases, the first two lasing modes are considered in Figs. 1(a) and 1(b), respectively. In order to obtain a quantitative measure of the similarity

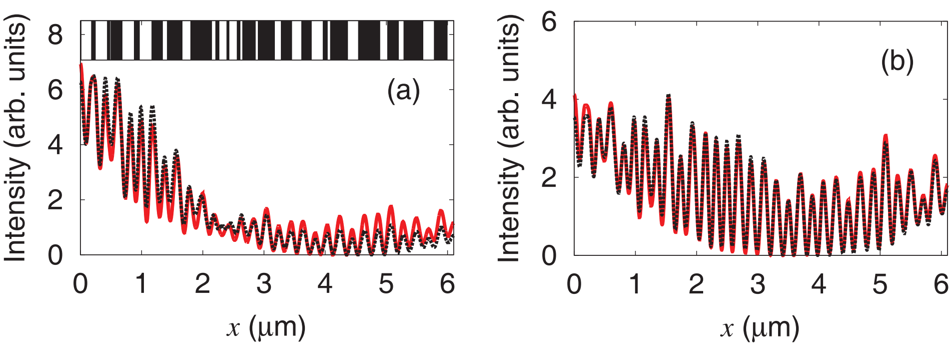

Fig. 1. (Color online) UNIFORM AND NONUNIFORM GAIN: Intensity distributions of the (a) first and (b) second threshold lasing modes; these are the first two modes to lase as the pumping rate increases. The lasing modes present with uniform gain (in both the scatterers and air gaps) are represented by solid red lines. The modes present with gain only in the air gaps by dashed black lines. The inset in (a) shows the locations of the air gaps (black bars). The differences due to nonuniform gain are (a) $D = 17\%$ and (b) $D = 6.7\%$. Although nonuniform gain can significantly modify lasing modes, this situation is not referred to as partial pumping throughout the paper. Instead, it is referred to as uniform pumping, as is frequently done, because gain is distributed uniformly throughout the random medium and is pumped at every location in which it is present.

of two intensity distributions $|E_i|^2(x)$ and $|E_j|^2(x)$, we use the difference

$$D_{ij} = \int_0^L ||E_i|^2(x) - |E_j|^2(x)|dx \times 100\%\,, \tag{5}$$

where the distributions $|E_{i,j}|^2(x)$ are normalized to one. The differences due to nonuniform gain are 17% and 6.7%, for the first and second lasing modes, respectively. In general, the precise value of D can vary drastically from mode to mode and depends on the realization of randomness.[42] However, in spite of these quantitative differences, the modes keep their main identity; for instance, they keep the same number of extrema and nodes. This is understandable as long as the imaginary part of the refractive index (due to optical gain) is small compared to the real part; in this case, the ratio of the imaginary to real parts is roughly 10^{-4}. Hence, in what follows, we will not consider this usual situation of nonuniform, but almost uniform (gain only in the scatterers or air gaps) gain as partial pumping. On the contrary, we will consider situations where the spatial extent of the gain does not cover the entire scattering sample, as when using a mask or focusing on the pump beam. We refer to this as intentional partial pumping and address it in the following sections.

2.3. *Intentional use of partial pumping*

One case of partial pumping to consider is incomplete penetration of the pump in a thick sample.[43] Then, only modes near the surface can be excited. In many cases, like in polycrystalline films or perforated membranes, this is not an issue. However, incomplete penetration of the pump was used in an initial experiment[44] by Varsanyi that anticipates the effects of partial pumping in random lasers in what he termed a "powder laser." By increasing the pump intensity I_0 on a solid-state sample, the depth of the pumped domain increases [see Eq. (1)] and changes the shape of the pumped area. At the lowest pump intensities, the output was directed along the surface of the sample perpendicularly to the pump beam. At higher pump intensities, when the depth of the pumped domain exceeded the transverse spot size on the sample, the laser entered "penetration mode" with output directed normal to the surface. Similar results have been seen in modern experiments, with confirmation of the role played by the scattering strength.[34] Materials used today have resulted in direct observation of the partially pumped active lasing area.[45]

Partial pumping was critical to the observation of a random laser with resonant feedback that was first realized in 1999 by Cao *et al.* using semiconductor powder[22] and a short time later in organic media.[26] As discussed in the previous section, the narrow spectral lasing lines arise from recurrent light scattering[46] resulting in the formation of spatial resonances. Irrespective of the scattering strength, the number of modes, localized and/or extended, increases quickly[47] with respect to the pump area in two- and three-dimensional random systems. The dependence of extended modes on the pump area is not trivial. Even in the absence of absorption, the pump can significantly modify lasing modes. Such behavior of extended modes and their dependence on the pump area shall be addressed throughout Sec. 4 and in more detail in Sec. 4.6. For now, it suffices to say that if the pump area is large enough, the narrow lasing lines overlap and the emission spectrum becomes smooth once again,[26] similar to the ASE regime. Hence, using small pump areas facilitated the experimental observation of narrow spectral lines in those pioneer experiments.[a]

The effect of the size of the excitation spot was recently studied experimentally in a porous GaP random laser[49] by changing the radius of the pump spot from 8.5 μm to 104 μm. For the small pump spot, the spectrum displayed clear, well separated, sharp peaks. For the large pump spot, the spectrum consisted of a wide peak, characteristic of ASE. However, the authors stress that in their experiment, ASE might also be due to a thin dye layer located at the sample surface. Hence, experimental conclusions about the effect of the size of the excitation spot are not yet definitive.

A few years earlier, the same group used spot pumping to study the spatial extent of random laser modes.[50] They tracked various lasing peaks in the emission spectrum as the position of a random sample of porous GaP was moved across the fixed partial pump spot. Spectral lasing peaks appeared, increased, then reduced in height and disappeared as a function of pump displacement on the sample. The spatial extent of the mode was defined by the difference in sample position where the peak appeared and disappeared. Mode extents measured in this way were found to be mostly on the same order of magnitude as the size of the pump spot. For large pump spots, lasing modes can occupy completely different spatial regions, but have the same frequency. Meanwhile, for small pump spots, like those observed, spectral repulsion occurs.[51] Indeed, the statistics in the experiment above indicated the presence of level repulsion.

Finally, the dependence of the lasing threshold on the size of the pumped area was an early concern of random lasers. As intuitively expected due to the reduction of gain, partial pumping has been shown to increase random laser thresholds.[52] The behavior of the threshold can be calculated based on the diffusion model[20] to follow the inverse square of the pump spot diameter.[53] However, it was found[54] that the specific behavior depends on the exact system under investigation, so that in some cases, the threshold depends on merely the inverse pump size. The main reason for the improved estimation was the boundary conditions on the pumped volume. It is therefore important to consider not only the actively pumped region of a partially pumped random laser, but also the unpumped region and the interface between the two. For example, such considerations have improved the efficiency to which the pump energy is used.[55] We shall examine, in the sections following, the spectral and spatial implications of partial pumping, where the unpumped region can play a strong role.

[a]The composition of the smooth, broad lasing peak for large pump areas is unknown, though some progress is being made.[24,48]

3. Spot Pumping in the Localized Regime

The general account of the different scattering regimes of a random laser, in Sec. 2.1, did not make any reference to partial pumping. Indeed, these general considerations implicitly assumed uniform pumping of the random medium. However, on the experimental side, uniform pumping is the exception rather than the rule, usually not for studying partial pumping *per se*, but for practical purposes such as pump enhancement through focusing of the beam. Typical pump beams for random lasers are focused onto a subregion of a random sample. Inhomogeneities in the spatial profile of pump intensity may exist in such a pump spot,[56] the implications of which, if controlled properly, shall later be shown to yield a powerful method of control. Here, however, we limit our exploration to consequences of a continuous and homogeneous pump spot. We shall review the theoretical and numerical work aimed at understanding the effect of local pumping. Consequences depend on the regime of wave propagation, i.e., diffusive or localized. We begin in this section with the localized regime, which is the most difficult to realize experimentally, but the easiest to analyze theoretically. We will address, in Sec. 4, the weakly scattering regime where partial pumping has more involved consequences.

Concurrent with the experimental work described in the previous section, analytical and numerical studies were devoted to partial pumping. As stated earlier, when several experiments in the late 1990s revealed narrow peaks in the emission spectrum on top of the photoluminescence band, it became clear that the diffusion model was insufficient to explain those new experimental observations. In conventional lasers the lasing peaks are known to be associated with the confined modes of the almost perfectly closed cavity. Hence, the idea was put forward that confined modes could also explain the observation of narrow lasing peaks in random lasers. However, in contrast with the usual behavior of a conventional laser, it was experimentally observed that the emission of a random laser changes with the size or the location of the pumping spot.

These two points, the existence of confined modes and the dependence of the laser emission on the pumping spot, have a natural explanation if scattering of light is strong enough to exhibit Anderson localization. For a random laser with lasing modes based on localized quasimodes, as discussed in Sec. 2.1, it is known that such states are confined and are equivalent to the cavity modes of a conventional laser. Now, the size of a localized mode typically ranges from one to several micrometers. Meanwhile, the experiments discussed in Sec. 2.3 utilize pump spot sizes that range from a few micrometers to millimeters. Therefore, the number of lasing modes will depend on the ratio of pump spot size to mode size. Furthermore, since localized modes usually reside at separate spatial locations, the modes that lase will depend on the position of the pump beam.

These predictions have been confirmed in numerical simulations of 2D random lasers.[38,39] First, consider a random system without gain which consists of a random collection of circular particles embedded in a background medium (Fig. 2).

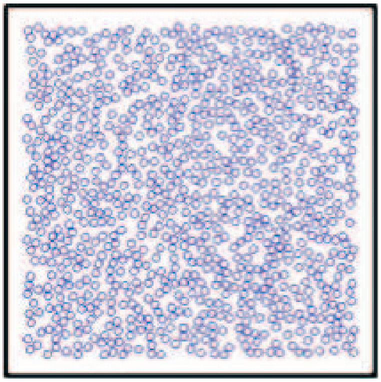

Fig. 2. (Color online) MULTIPLE-SCATTERING SYSTEM: Example of a random realization of circular scatterers contained in a square box of size $L = 5.5$ μm and background optical index $n = 1$. The radius and the optical index of the scatterers are, respectively, $r = 60$ nm and $n = 2$. The numerical grid is bounded by perfectly matched layers (not shown) to simulate an open system. When optical gain is added to this random collection of scatterers, a random laser can be formed.

To simulate an open system, perfectly matched layers are introduced at the boundaries of the numerical grid.[57] There is no absorption, neither in the scatterers nor in the background medium. A large index contrast is assigned between the scatterers and the background medium in order to reach the regime of Anderson localization. To check that this regime is reached, the time-dependent Maxwell's equations are solved using the finite-difference time-dependent (FDTD) method.[58] After launching a short pulse in this system, the time response is Fourier transformed in order to obtain the spectrum of eigenfrequencies. Next, the eigenmodes are excited individually by a monochromatic source. Examples of such eigenmodes of the system without gain are displayed in Fig. 3. They demonstrate that this system contains confined modes due to Anderson localization.

The corresponding 2D random laser is the same system for which the background medium becomes the gain medium, the optical index remaining unchanged. As first proposed by Jiang and Soukoulis in 1D systems,[59] the gain medium consists of four-level atoms that are uniformly distributed in the system with the exception of the high index scatterers. The time-dependent Maxwell's equations are now coupled with the polarization and population equations of the four-level atoms.

The results of a simple example of partial pumping are presented in Fig. 4. In the left panel of Fig. 4, only the left half of the system is pumped and in the right panel, only the right half is pumped. The spectra are very different and demonstrate that the modes excited depend on the pump position. This is in agreement with experimental observations.

Another important result due to using local pumping is that the lasing modes obtained by the full time-dependent model with gain have been found to be identical with a good precision to localized states of the corresponding passive system without

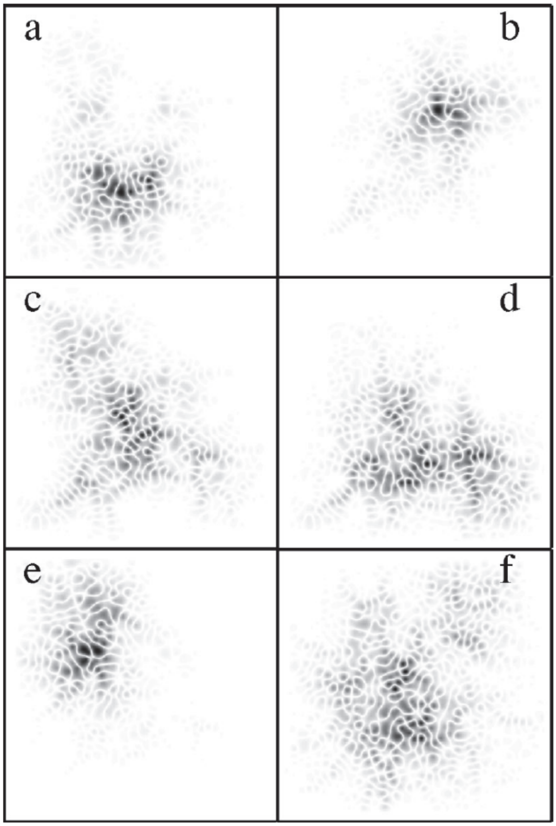

Fig. 3. LOCALIZED EIGENMODES: Spatial distribution of the field amplitude of six localized modes of the passive system shown in Fig. 2. The existence of such eigenmodes indicates that the scattering regime is that of Anderson localization.

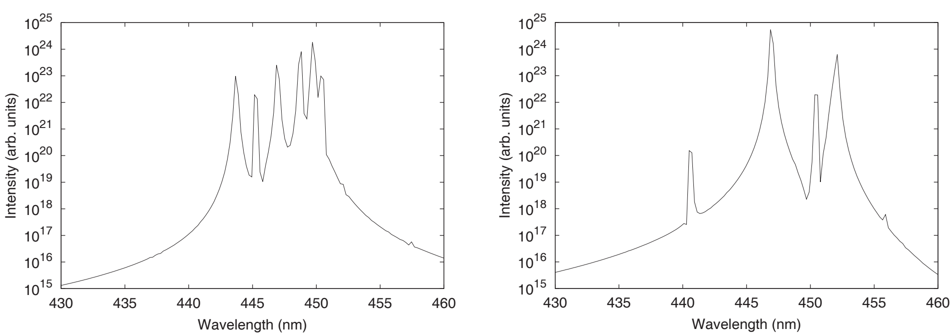

Fig. 4. SPATIALLY DEPENDENT PARTIAL PUMPING EMISSION: Frequency spectra of the 2D random laser when pumping only (left panel) the left half of the system and (right panel) the right half. Different locations of the pump are seen to probe different realizations of disorder.

gain.[38,39] Similar results were obtained in 1D systems by Jiang and Soukoulis,[40] thus confirming the part played by the confined modes of strongly scattering random media.

To make such a comparison, the system had to be in the single mode regime in order to compare the single lasing mode to the corresponding localized mode of the passive system. To do this, one adjusts the pumping rate near the lasing threshold so that only one mode is excited. The result is to excite the most favored mode, i.e., the threshold lasing mode. If the pumping rate W_p is progressively increased, the number of lasing modes increases each time W_p crosses the threshold for the next lasing mode. Then, it appears it is not possible to individually excite lasing modes other than the threshold lasing mode, making a comparison of higher-threshold lasing modes with the localized modes of the passive random system difficult.

The reasoning above implicitly assumes uniform pumping of the gain medium. However, it appears possible to take advantage of the fact that the modes are localized and are usually positioned at different locations of the random medium. It was then proposed to pump the system locally in order to excite the mode located at the position of the pump.[38,39]

Examples of local excitation are displayed in Fig. 5, where the circle indicates the location of the pump spot. The pump has a Gaussian spatial profile of width ~ 0.5 μm, which is close to the value of the localization length in this system. The spatial maps of the laser modes are identical to the localized modes d and e

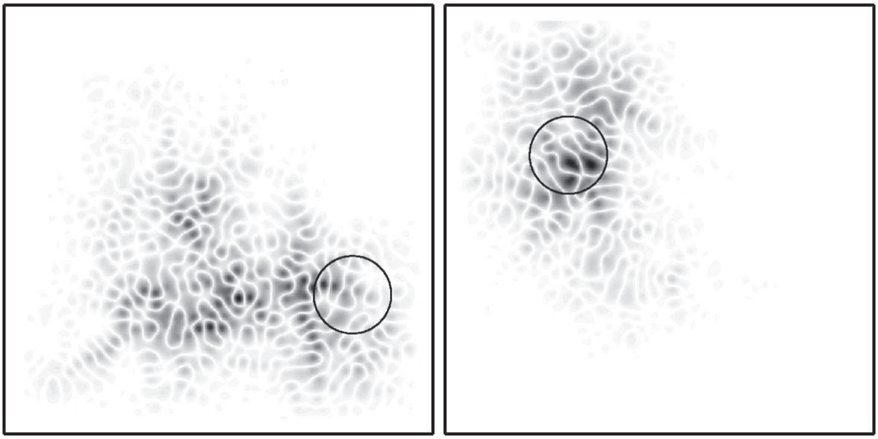

Fig. 5. LOCALIZED MODE EXCITATION WITH PARTIAL PUMPING: Spatial distribution of the field amplitude for spot pumping. The circles represent the spatial extension of the Gaussian pump of width ~ 0.5 μm. Compared with the field distribution of (left panel) mode d and (right panel) mode e in Fig. 3, these lasing modes are very similar. The modes here are localized and have large quality factors. Therefore, the pump intensity does not need to be large in order to excite them. Subsequently, although pumping is only partial, the low pump intensity does not significantly alter the character of the lasing mode, especially since absorption does not exist in the unpumped region.

of the same system without gain (Fig. 3). Such results were obtained by properly choosing the position and the rate of the pump. It was confirmed that the modes of the passive random system act as ordinary modes of a conventional laser cavity. It is also important to stress that the most favored mode of this system is mode b in Fig. 3. Hence, without local pumping it would have been impossible to excite modes d and e individually.

Although it is experimentally possible to focus the pump spot size to small areas, the positions of localized lasing modes in the sample are unknown *a priori*. This makes it difficult to position the pump beam for individual mode selection. One suggestion[60] is that a defect be introduced that precisely determines the location of the lasing mode. Numerous other techniques that rely on manipulation of the underlying random structure[61–69] have also been suggested in order to obtain control over random lasers. However, we shall see later that the pump alone can be used to exercise strong influence over random laser emission.

4. Spot Pumping in the Weakly Scattering Regime

The weakly scattering regime here refers to systems that may be diffusive or ballistic. In contrast to the localized regime, the modes or resonances are extended across the random system and, therefore, experience strong leakage through the external boundaries. Because of these two features, the effect of partial pumping will differ from the localized case. We will see that spot pumping significantly modifies the spatial and spectral features of lasing modes.

First, two different situations must be distinguished depending on the presence or absence of absorption of the laser field. For instance, laser dyes can exhibit overlapping gain and absorption spectra, so that any emission produced within a partially pumped active area of a random laser can be reabsorbed once it exits into the unpumped region. Without absorption, the amplitude of the field outside the gain region, where there is no amplification, is determined by the total field amplitude at the gain boundary. This intensity can be large and scattering in the unpumped region can produce feedback into the pumped area. With absorption, any such effects are suppressed, and in diffusive random lasers, it was found to induce confinement of random laser modes.

In this section, we review recent numerical work devoted to partial pumping in 1D and quasi-1D weakly scattering systems. After the presentation of the systems and of the numerical methods, we examine how the spectral and spatial properties of lasing modes depend on the extension of the pumped domain. We conclude the section with an experimental demonstration of a partially pumped quasi-1D random laser.

4.1. *Numerical methods*

One of the primary methods used to generate results in this review is frequency based and relies on the transfer matrix;[70,71] all results in this case are from one-dimensional (1D) systems. Optical gain is simulated via a complex refractive index. Another significant tool is based on the FDTD method.[58] In that case, gain can be simulated by a negative conductance or by considering the populations and polarization of actual multilevel atoms, which are then coupled to the light field. Results from FDTD methods are also presented in this review. However, many references exist that describe such methods in depth.[24,39,72–75] Therefore, we present here only the transfer matrix method for convenience.

The 1D random systems are composed of N dielectric layers, half with index of refraction n_1 alternating with air gaps ($n_2 = 1$) resulting in a spatially modulated index of refraction $n(x)$. The system is randomized by specifying different thicknesses for each of the layers as $d_{1,2} = \langle d_{1,2}\rangle(1 + \eta\zeta)$, where $\langle d_1\rangle$ and $\langle d_2\rangle$ are the average thicknesses of the layers, $0 < \eta < 1$ represents the degree of randomness, and ζ is a random number in $(-1, 1)$.

This method is used to calculate both the quasimodes and the lasing modes of a random structure. Electric fields on the left (right) side of the structure $p_0(q_N)$ and $q_0(p_N)$ travel toward and away from the structure, respectively. Propagation through the structure is calculated via the 2×2 matrix M,

$$\begin{pmatrix} p_N \\ q_N \end{pmatrix} = M \begin{pmatrix} p_0 \\ q_0 \end{pmatrix}. \tag{6}$$

The boundary conditions for outgoing fields only are $p_0 = q_N = 0$, requiring $M_{22} = 0$ for the solutions. Verification of solutions (with or without gain) is provided by the phase of M_{22}, calculated as $\theta = \mathrm{atan2}(\mathrm{Im}M_{22}, \mathrm{Re}M_{22})$. Locations of vanishing M_{22} give rise to phase singularities since both the real and imaginary parts of M_{22} vanish. The phase change around a path surrounding a singularity in units of 2π is referred to as topological charge,[76,77] which has the values $+1$ and -1 for the cases studied.

4.2. *Spectral behavior*

In this section, we see how partial pumping affects lasing frequency behavior. $N = 161$ layers are used, with $\eta = 0.9$, $n_1 = 1.05$, $\langle d_1\rangle = 100$ nm, and $\langle d_2\rangle = 200$ nm, resulting in a total average length of $\langle L\rangle = 24{,}100$ nm. Linear gain is simulated by appending an imaginary part to the index of refraction $\tilde{n}(x) = n_r(x) + in_i$, where $n_i < 0$. The value of n_i is first chosen constant everywhere within the random system. Next, nonuniform gain is introduced by multiplying n_i by a step function $f_E(x) = H(-x + l_G)$, where $x = 0$ is the left edge of the random structure and $x = l_G$ is the location of the right edge of the gain region. l_G may be chosen as any value between 0 and L.

Figure 6 maps the wavelength and threshold values of lasing modes as the pump spot length from uniform pumping ($l_G = L = 24{,}100$ nm) reduces to that of partial pumping ($l_G < L$). As the size of the pump changes, we will see in Sec. 4.4 that the envelopes of the intensity distributions change, but for most modes, the amount of

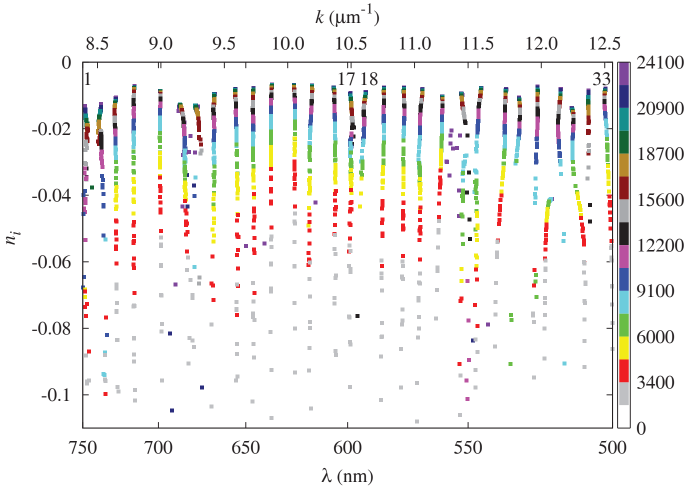

Fig. 6. (Color online) LASING MODE BEHAVIOR WITH PUMP SPOT SIZE: Wavelengths (given by $\lambda = 2\pi/k$) and thresholds (n_i) of the lasing modes of a 1D random laser. Four lasing modes 1, 17, 18 and 33 are explicitly marked. The pump spot length l_G reduces from uniform pumping ($l_G = L$) to partial pumping $l_G < L$. The color indicates the value of l_G (units of nm) decremented along the scatterer interfaces. As the pump spot size reduces, the number of modes reduces and they become more spectrally separated.

gain (n_i) is small enough to leave the optical index unchanged. Thus, as the size of the pumped region changes, their frequencies remain roughly the same as in the uniform pumping case. Similar behavior of lasing mode frequencies can be seen as the pump length is varied in a simpler cavity with uniform index. Thus, the robustness of frequency is not due to inhomogeneity in the spatial dielectric function, but to the robustness of the optical index landscape. However, the threshold values of the lasing modes increase as l_G decreases, due to the limited spatial region of amplification.

4.3. *Creation of new lasing modes*

Figure 6 shows that some modes stop lasing, e.g., mode 18, no matter how large the gain. It was found[71] that lasing modes not only disappear, but can be created.[78] The new lasing modes typically exist for specific sizes of the pump spot and disappear as the pump spot is altered. They appear at various frequencies for several different pump spot configurations.

New lasing modes, as reported, are always created with larger thresholds than the existing lasing modes adjacent in frequency. The disappearance of lasing modes is not caused by mode competition for gain, because gain saturation is not included in this model of *linear gain*. Disappearance/appearance events occur more frequently for smaller pump spots. It is also found that the disappearance events can exhibit behavioral symmetry (as explained below) around particular values of

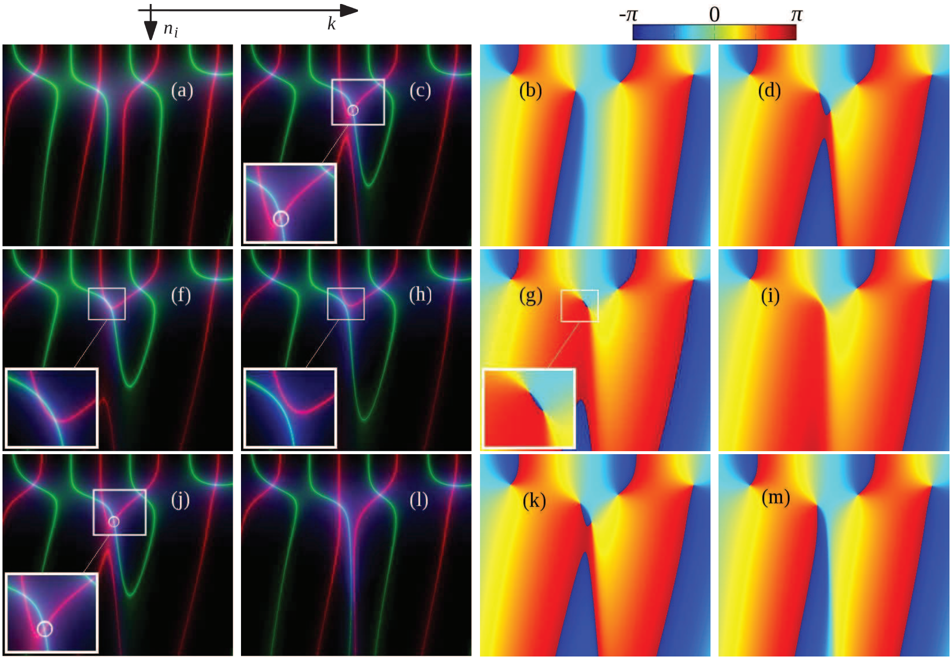

Fig. 7. (Color online) LASING MODE CREATION AND ANNIHILATION WITH PUMP SPOT SIZE: (left) Red lines represent real and green lines represent imaginary zero lines of the transfer matrix element M_{22}. Their crossings indicate (k, n_i) values of lasing modes. (right) Phase maps of M_{22} validate lasing mode solutions. All data are in the ranges (10.3 μm^{-1} < k < 10.8 μm^{-1}) and ($0 \geq n_i \geq -0.074$) covering lasing modes 17 and 18 for pump spot size l_G = 14961 nm [(a) and (b)], 14,553 nm [(c) and (d)], 14,523 nm [(f) and (g)], 14,472 nm [(h) and (i)], 14,284 nm [(j) and (k)] and 14,042 nm [(l) and (m)]. The joining of zero lines in (c) results in the formation of a new lasing mode (new zero line crossing is encircled in white). The inset in (c) is an enlargement of the mode 17 and new mode solutions. Such creation/annihilation events are common as the pump spot size is changed. This results in stochastic spectral behavior, but in general, the number of modes reduces as the pump spot size reduces.

l_G. This disappearance and subsequent reappearance causes a fluctuation of the local density of lasing states as l_G changes. Therefore, although the individual laser mode frequencies are generally robust, their existence is not. This means the total spectral emission can still, generally, be sensitive to the pump spot.

The progression of one representative event is examined in detail. Lasing mode 17 does not exist for pump spot lengths in the range $10,500$ nm $\leq l_G \leq 14,500$ nm. Figure 7 shows the real and imaginary zero lines of the transfer matrix element M_{22} and their accompanying phase maps for pump spot sizes in this range. A value of $\mathrm{Re}M_{22} = \mathrm{Im}M_{22} = 0$ indicates outgoing boundary conditions are satisfied and that a lasing solution is found. At this location, a singularity appears in the phase map. As l_G decreases, the zero lines of lasing modes 17 and 18 join as seen in the transition from Figs. 7(a) to 7(c). This creates a new mode solution (marked by a white circle) with a frequency between lasing modes 17 and 18 and a larger threshold. The existence of a new lasing mode is confirmed by the phase singularity in Fig. 7(d). The new mode is close to mode 17 in the (k, n_i) plane and its phase singularity has the opposite topological charge as seen in Fig. 7(d). As l_G decreases further, the joined zero lines forming mode 17 and the new mode pull apart. This causes the two solutions to approach each other in the (k, n_i) plane, i.e., the frequency and threshold of mode 17 increase while the frequency and threshold of the new mode decrease. In Figs. 7(f) and 7(g), the solutions are so close that they are nearly identical, yet they still represent two separate solutions. Further decreasing l_G makes the solutions identical. The zero lines then separate and the phase singularities of opposite charge annihilate each other in Figs. 7(h) and 7(i). This results in the disappearance of mode 17 and the new mode. The process then reverses itself as l_G is decreased further [Figs. 7(j)–7(m)] yielding the reappearance of mode 17 and the new mode and their subsequent separation in the (k, n_i) plane. This is the aforementioned behavioral symmetry around $l_G = 14,472$ nm.

The existence of new lasing modes in the presence of gain saturation has been confirmed[78] through full-wave FDTD calculations. Three lasing modes were investigated, two original modes and one newly created mode, using the same parameters as in the transfer matrix method above. However, the gain curve was narrowed in order to excite only one mode and exclude all others. For example, when the gain curve is centered on the lasing frequency of the newly created mode, it is the first to lase instead of the original modes, but its absolute lasing threshold is higher. In this manner, the thresholds could be calculated and compared to those from the transfer matrix method. Nearly identical behavior was found, indicating such modes are robust against the introduction of more realistic conditions, such as spatial hole burning and mode interaction.

The newly created modes were also found to exist in simple optical cavities with a uniform index of refraction.[79] Thus, this phenomenon is not limited to random lasers. The mechanism for their creation stems from the gain boundary introduced by partial pumping and feedback from the unpumped region. These modes tend to obtain maximum intensity at the gain boundary, so have been referred to as surface modes. They therefore have great directionality and may find potential applications in optical switching and sensing.

4.4. *Spatial properties of modes*

Lasing with coherent feedback in weakly scattering systems requires focusing of pump light in order to observe discrete lasing peaks.[26,47,81] Imaging of light on the sample surface revealed that the lasing modes were spatially restricted to the pumped domain, not extended over the scattering medium as expected. This result was puzzling because, as discussed in Sec. 2.1, lasing is expected to use underlying quasimodes of the random system, which are not localized in the presence of weak scattering. It was shown,[80] however, that in the presence of absorption, lasing need not occur strictly in the quasimodes of the entire passive random medium. Instead,

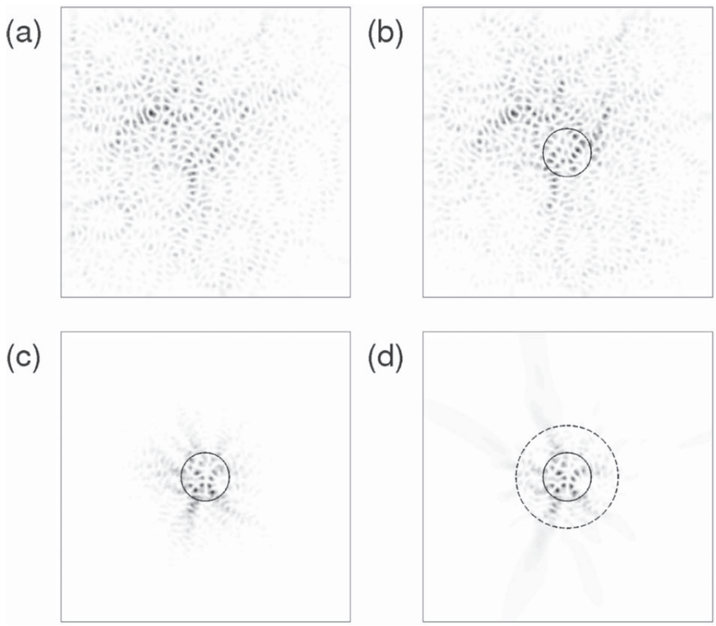

Fig. 8. ABSORPTION-INDUCED MODE CONFINEMENT: Spatial intensity distribution of (a) the quasimode with the longest lifetime in a passive diffusive 2D random medium, (b) the first lasing mode excited by partially pumping inside the circular region near the center and no absorption outside it, (c) the first lasing mode with absorption outside the circular region, (d) the first lasing mode with the scatterers beyond one absorption length (dashed circle) completely removed. Absorption suppresses feedback beyond the pumped region, thereby confining the lasing mode to a smaller spatial region than is otherwise possible in a weakly scattering system. Reprinted with permission.[80]

reabsorption of emitted light from the pumped domain suppresses feedback in the unpumped region. Lasing modes are therefore formed anew and rely on quasimodes of the underlying *reduced* system. This was explored experimentally by Wu *et al.*[34]

The type of reabsorption described above can occur, for example, when the gain and absorption spectra overlap. Illustrated in Fig. 8, a "cavity" is induced with boundaries generated between the gain region formed by pumping and the absorption region outside. Compared to the case without absorption [Fig. 8(b)], feedback is suppressed when absorption is present [Fig. 8(c)], effectively reducing the size of the system. This was verified by removing all of the scatterers outside the partially pumped region (including one absorption length) in Fig. 8(d). The total number of possible lasing modes are essentially dictated by the size of this absorption-induced cavity. The size of the modes, also dictated by the effective cavity, approaches the size of the pump spot. Therefore, for very small pump spots, the lasing mode size may approach that of a localized mode. Keep in mind, however, that the mode of this underlying passive system is spatially extended. The quasimode of the entire medium is shown in Fig. 8(a). In the reduced system of Fig. 8(d),

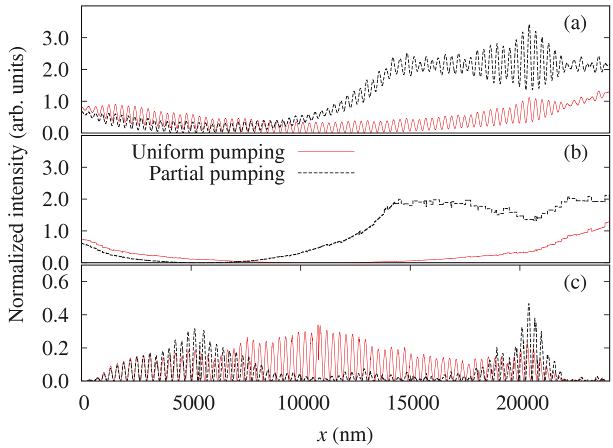

Fig. 9. (Color online) PUMP SPOT ALTERS LASING MODES WITHOUT ABSORPTION: Normalized intensity of a single extended lasing mode. The mode in the presence of uniform pumping is represented by solid red lines and with partial pumping by dashed black lines. In this case, only the spatial range $0 \leq x \leq 14,000$ nm is pumped. This single mode is presented in terms of its: (a) total intensity, (b) traveling wave intensity, and (c) standing wave intensity. Partial pumping significantly changes the spatial intensity envelope as well as the standing wave and traveling wave components. These alterations take place even without the presence of absorption in the unpumped region.

without gain present, the mode will be extended across the reduced cavity and have a large leakage rate due to weak scattering (and few number of scatterers).

The situation changes if no absorption exists in the unpumped region. Feedback is no longer suppressed, but the pump spot maintains a significant influence over the lasing modes. An extended mode in the 1D system considered in Secs. 4.2 and 4.3 is illustrated in Fig. 9(a). Amplification is high due to the large amount of gain required to compensate for the leakage through the open boundaries. With uniform as with partial pumping, this results in a buildup of intensity along the outer gain boundary of the pumped region.[29,71] Such intensity profiles are characteristic of leaky systems and are also observed, for instance, in distributed feedback (DFB) lasers.[82]

Figure 9(a) shows that the spatial profile of lasing modes also changes significantly by partial pumping, even without absorption in the unpumped region, in agreement with experimental observations.[34,83] As a result of leakage of energy through the open boundaries of the system, the extended mode contains a noticeable traveling wave component. This is in contrast to confined modes in a closed cavity or localized modes, which are standing waves to a good approximation. The standing and traveling components are defined[71] as follows.[b] At every spatial location x, the right-going complex field $\Psi^{(R)}(x)$ and left-going complex field $\Psi^{(L)}(x)$

[b]Flux conservation is not required in the presence of gain.[84]

are compared. The field with the smaller amplitude is used for the standing wave and the remainder for the traveling wave. In other words, if $|\Psi^{(R)}(x)| < |\Psi^{(L)}(x)|$, then the standing wave $\Psi^{(S)}(x)$ and traveling wave $\Psi^{(T)}(x)$ components are

$$\Psi^{(S)}(x) = \Psi^{(R)}(x) + [\Psi^{(R)}(x)]^* \,, \tag{7a}$$

$$\Psi^{(T)}(x) = \Psi^{(L)}(x) - [\Psi^{(R)}(x)]^* \,. \tag{7b}$$

The traveling wave component directed out of the random sample is displayed in Fig. 9(b). With uniform pumping, the traveling component has maxima at the boundaries of the system, where the field intensity is maximum and energy flows out of the system. In the case of partial pumping, the traveling wave component has maxima at the boundaries of the gain region and in the unpumped region.

The standing wave component is displayed in Fig. 9(c). In the case of uniform pumping, the standing wave component is at its maximum at the center of the system, which is also the center of the gain domain. This is the location where the traveling wave component is at a minimum, i.e., where the two components traveling to the left and right compensate each other. In the case of partial pumping, the standing wave component has a maximum at the center of the pumped region, where the traveling wave component is also at a minimum.

This behavior is well illustrated by introducing the "center" of an extended lasing mode. The center is defined as the location x where the relative strength of the standing wave is at a maximum. The relative strength is calculated through the ratio of standing wave amplitude to traveling wave amplitude. As just discussed, where

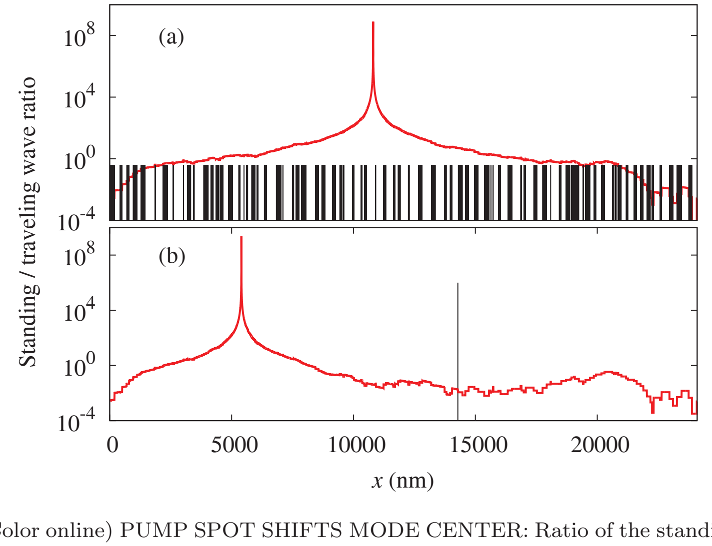

Fig. 10. (Color online) PUMP SPOT SHIFTS MODE CENTER: Ratio of the standing/traveling wave components of a single lasing mode in a 1D random laser. (a) Uniform pumping is present while in (b) partial pumping exists in the range $0 \leq x \leq 14,000$ nm. The random structure is shown in the lower portion of (a). A vertical line marks the right edge of the pump spot in (b). This low-threshold mode moves its center along with the pump spot.

the traveling waves moving to the left and to the right are balanced, the standing wave is largest, the traveling component is minimum, and the ratio of the two diverges. Results from considering uniform and partial pumping for a representative lasing mode are shown in Fig. 10. The lasing mode center is located near the center of the total system when considering uniform pumping in Fig. 10(a). With the size of the pump spot reduced in Fig. 10(b), we see that the lasing mode center moves to stay near the middle of the pumped region. More generally, it has been observed[71] that the center of an extended lasing mode can follow the location of the pump spot. It has also been found that in general, the modes most likely to lase (i.e., the ones with the lowest lasing thresholds) are those that can move their mode centers near the center of the pumped region.

As shown in Fig. 9(c), the standing wave component has a second maximum that is located in the unpumped region. This maximum can be attributed to wave scattering and resonance in the "local cavity" formed by the unpumped region. However, unlike the mode center, there is a significant traveling component at the location of this second maximum.

4.5. *Quasimode mixing*

Far above the lasing threshold, it was found that lasing modes consist of a collection of states formed by the random system without gain.[29] Mode mixing in this regime is largely determined by nonlinear effects from gain saturation. However partial pumping alone can also cause mode mixing.[71] In this case, lasing modes are decomposed in terms of quasimodes and found to be a superposition of those nearest in frequency. The more pumping deviates from being uniform, the larger the contribution from nearby quasimodes.

Figure 11(a) shows the decomposition of a lasing mode with uniform and partial pumping. Beginning with the case of uniform gain ($l_G = L = 24,100$ nm), the largest contribution to lasing mode 17 is from corresponding quasimode 17. There is a nonzero contribution from other quasimodes on the order 10^{-3}. This reflects slight differences between the lasing mode profile in the presence of uniform gain and the quasimode profile.[70,85] With the pump spot length reduced to $l_G = 14,284$ nm, the coefficients from quasimodes closer in frequency to the lasing modes increase significantly, i.e., quasimode mixing occurs. The exceptions are the very leaky quasimodes 7, 14 and 23. Unlike lasing mode 17, quasimodes 7, 14 and 23 have intensities which are peaked at the opposite boundary of the structure, making their overlap minimal. As the pump spot size reduces, lasing mode 17 is tipped further in the opposite direction, reducing overlap with these modes even more.

Figure 11(b) reveals the five largest coefficients as the pump spot size is incrementally reduced along the scatterer interfaces. While the lasing mode remains dominantly composed of its corresponding quasimode, neighboring quasimodes mix in significantly. It is known that linear contributions from gain induced polarization bring about a coupling between quasimodes of the passive system.[86] However, this

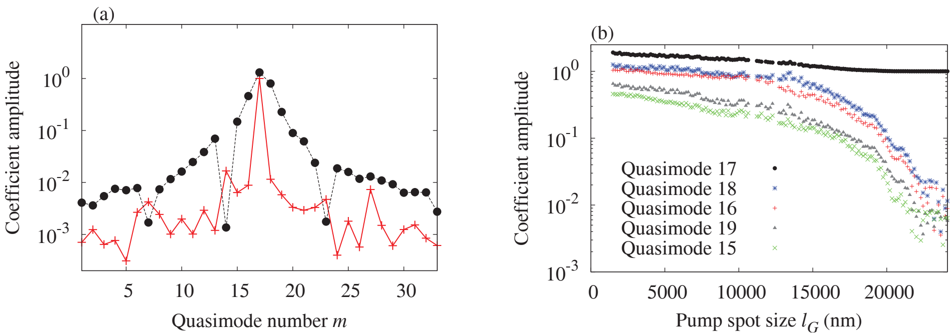

Fig. 11. (Color online) PUMP SIZE DETERMINES MODE MIXING: Decomposition of lasing mode 17 in terms of quasimodes of the total passive system. (a) Decomposition with (red crosses) uniform pumping and (black circles) partial pumping. (b) Five largest coefficients from the decomposition of lasing mode 17. Some coefficients are greater than one, which is possible in open systems. No absorption exists in this system. As the pump spot size l_G is reduced, the amount of mode mixing increases dramatically.

effect is small compared to the mode mixing caused by partial pumping, as observed in Fig. 11(b).

4.6. *Threshold statistics*

We have often discussed the apparent requirement of partial pumping in observing discrete, narrow lasing peaks in large samples, especially when scattering is weak. The most commonly cited reason for this behavior is that smaller pump areas excite fewer modes so that individual narrow peaks become distinguishable in the spectrum. This is certainly the case when absorption is present in the unpumped region. We discussed earlier that with absorption, lasing mode properties are dictated by the effective system size, given by the pumped region plus the absorption length (see Sec. 4.4). In this case, spectral behavior of random lasers with respect to pump size is similar to spectral behavior of passive random systems with respect to the system size. Since the number of resonances reduces with the system size, the number of lasing modes also reduces with the size of the pumped region. Hence, the frequency spacing of lasing modes increases.

However, there are cases where the above argument does not hold. First, absorption is not always present and it was shown above that scattering in the unpumped region can have a large influence on random lasers so that the system cannot be considered limited to the pumped domain. Next, in weakly scattering systems, a narrow distribution of decay rates exists, due to reduced scattering.[87,88] The distribution (normalized by the average decay rate) gets even narrower as the system size decreases,[89,90] suggesting strong competition among very similar modes. Thus, selecting individual modes for lasing in this regime using partial pumping would seem to be more difficult. However, experiments[26,34,47] consistently show the observation of discrete lasing peaks when the pumped region is small enough.

How can this apparent contradiction be explained? First, due to mode mixing, inhomegeneity of the dielectric function, and the creation and destruction of lasing modes, a one-to-one correspondence between lasing modes of the partially pumped system and those of the uniformly pumped system (or resonances of the passive system) does not exist. Thus, a partially pumped random laser is expected to behave differently from predictions based on the passive random system.[91,92]

Next, a statistical study of lasing thresholds is presented, which shows that partial pumping leads to an increase of the width of their distribution. The result is that for a given value of the gain, there are less lasing modes with partial pumping than with uniform pumping.

Considering $10,000$ realizations of 1D random structures, lasing thresholds (n_i) were studied[93] statistically using the transfer matrix method. $N = 41$ layers are used, with $\eta = 0.9$, $n_1 = 1.05$, $\langle d_1 \rangle = 100$ nm, and $\langle d_2 \rangle = 200$ nm, resulting in a total average length of $\langle L \rangle = 6100$ nm. Figure 12 shows the distribution of thresholds $P(n_i)$ for uniform and partial pumping; no absorption is included for partial pumping. A large-threshold tail is observed past a kink at $n_i \sim 0.175$ in the partial pumping case. The reason for the sharp kink between small and large-threshold modes is due to the newly created, high-threshold modes discussed in the previous section. This tail highly distorts the threshold statistics which is evident in the skewness S that characterizes the degree of asymmetry around the mean value. The skewness increases from $S = 1.4$ for uniform pumping to $S = 2.2$ for partial pumping.

Large-threshold modes rarely lase experimentally, so only $n_i \leq 0.175$ is considered with partial pumping and the distribution is re-normalized, shown in the inset in Fig. 12 (the distribution for uniform pumping is left unchanged). Due to asymmetry (even uniform pumping has $S > 1$), the first moment of the distribution is

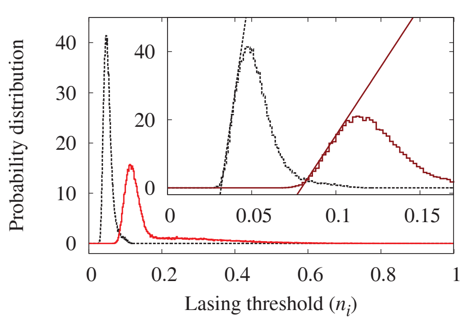

Fig. 12. (Color online) THRESHOLD DISTRIBUTION WITH PARTIAL PUMPING: Probability distributions of lasing thresholds for (dashed black lines) uniform pumping and (solid red lines) partial pumping. The inset shows the *re-normalized* probability distribution (solid dark-red lines) for partial pumping. The rising slope of the first main peak is 4.5 times greater with uniform pumping than with partial pumping. This illustrates how slow adjustments of the pump rate result in a greater number of modes being excited with uniform pumping as opposed to partial pumping.

characterized using the most probable threshold n_m rather than the mean threshold $\langle n_i \rangle$. n_m shifts from 0.047 for uniform pumping to 0.112 for partial pumping, a factor of 2 increase. The standard deviation around n_m increases from $\sigma = 0.012$ for uniform pumping to $\sigma = 0.023$ for partial pumping, nearly twice as large. This indicates the fluctuation of thresholds increases for smaller pumping sizes. Furthermore, the inset of Fig. 12 shows the slope of the rising part of the re-normalized threshold distribution with partial pumping. The number of lasing modes dN_l within a threshold range dn_i is proportional to the slope m ($dN_l = m\ dn_i$). With partial pumping, m is 4.5 times smaller than with uniform pumping (including the large-threshold tail for partial pumping gives a slope 6 times smaller). If the pumping rate is gradually increased from zero, the number of available lasing modes can therefore be substantially less with partial pumping.

Figure 13 compares the threshold statistics for partial pumping ($\ell_G/L = 1/3$) with and without absorption in the unpumped region. The large-threshold modes have completely disappeared by adding absorption. The large-threshold tail without absorption has been excluded in order to compare the distributions directly. As mentioned earlier, the newly created large-threshold modes are spatially concentrated at the gain boundary, so that absorption can effectively kill these modes. Absorption also increases thresholds to $n_m = 0.129$, which is 15% larger than the absorptionless case.

With the partial pump spot three times less than the uniform pumping case, the number of modes is 67% less when absorption is present. Without absorption, the total number of modes with partial pumping is only 14–17% less than with uniform pumping. With the number of available lasing modes reduced, the frequency spacing

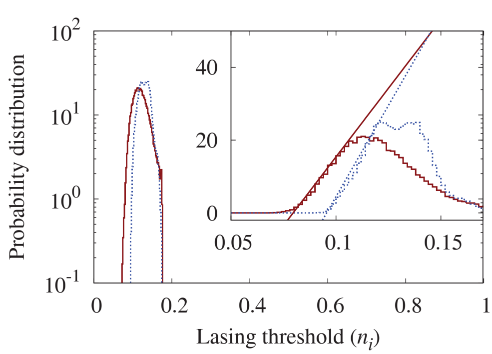

Fig. 13. (Color online) THRESHOLD DISTRIBUTION WITH ABSORPTION: Probability distributions of lasing thresholds for partial pumping without absorption (solid dark-red lines) and with absorption (dotted blue lines). The distribution without absorption excludes large-threshold modes, which are rarely excited, and has been re-normalized. Absorption clearly increases lasing thresholds, as expected. However, the inset also shows the rising slope is roughly 1.3 times greater with absorption than without absorption. This means that the number of lasing modes excited incrementally is greater with absorption present, making it more difficult to observe the threshold for single modes as the pumping rate is increased.

between them increases, however, we already saw that the mechanism is different than in the case with absorption.

The inset of Fig. 13 shows the slope of the rising part of the re-normalized distribution. Without absorption, the slope is 1.3 times smaller. The explanation of how the smaller slope leads to more easily observing lasing peaks in the emission spectrum is the same as discussed earlier, for Fig. 12. To elaborate, consider an experimentally obtained threshold pumping rate P_t and the experimentally limited pump step (e.g., by power fluctuations) ΔP. The relative experimental pump step is $\delta P = \Delta P/P_t$. A larger threshold means smaller allowable adjustments δP, thereby allowing a finer tuning of the pumping rate. The most probable, numerically obtained threshold n_m, gives a good representation of P_t. Thus, the relative pump step $\delta n_i = dn_i/n_m$ observed, may allow a finer tuning of the pumping rate, thereby making it easier to see modes begin lasing incrementally.

4.7. *Spot location and size*

Spectral changes of random laser emission under location changes of the pump area have been observed in various systems.[22,26,50] Recently, the spectral dependence of lasing emission on partial pumping in a quasi-1D microfluidic random laser was investigated experimentally.[94] In this section, we focus on this quasi-1D system and discuss the dramatic changes to the emission spectrum with respect to the size and location of the pump field. As the pump spot location is displaced or its size changed, different lasing modes are excited. This leads to the possible generation of many unique lasers within a single microfluidic system and bears potential interest for sensing applications.

The laser chip is made by soft photolithography of a negative photoresist mold followed by replica-molding of polymeric structures.[95] Eventually the polymeric imprint is plasma-bonded on a glass slide. Rhodamine 6G in ethanolic solution is then injected into the microfluidic channel (MFC) to serve as the gain medium. The dye circulation within the MFC allows flow-regeneration and eliminates dye bleaching. The initial mask shown in Fig. 14(a) is periodically structured with 10 μm $\times$ 20 μm rectangular pegs placed along the 3 mm length of the channel, separated from each other by 10 μm. After photolithography, we obtain a snake-shaped structure shown in Fig. 14(b). The limited accuracy of the fabrication process results in fluctuations in the layer thickness (10 μm $\pm$ 0.65 μm) significant enough at the optical scale to give rise to multiple scattering of light emitted by the dye molecules.

A Q-switch Nd:YAG laser is used at 532 nm to pump the dye in the MFC. A $f =$ 50 mm cylindrical lens focuses the pump beam into a 3 mm long, 10 μm wide stripe. The uniform illumination along the length of the MFC [highlighted in Fig. 14(b)] forces 1D random lasing. Emission spectra are recorded with a HR4000 (Ocean Optics) fiber-probe spectrometer having a spectral resolution of 0.11 nm. The MFC is imaged with the help of a Zeiss Axioexaminer microscope and a Hamamatsu Orca-R2 silicon CCD camera.

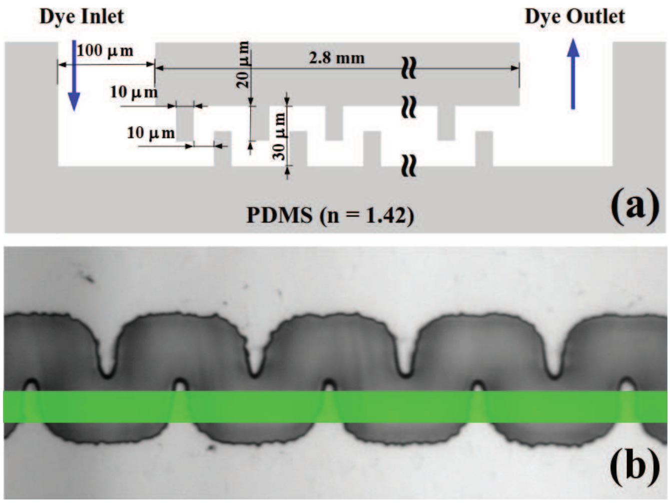

Fig. 14. (Color online) MICROFLUIDIC RANDOM LASER: (a) Schematic top-view of the 3 mm long MFC with 10 μm thick PDMS walls positioned periodically along its length with a 40 μm period. (b) An optical microscope image of the fabricated MFC showing the fluctuations of the width of each PDMS peg. The green stripe represents the pumping scheme used for performing random laser emission studies.

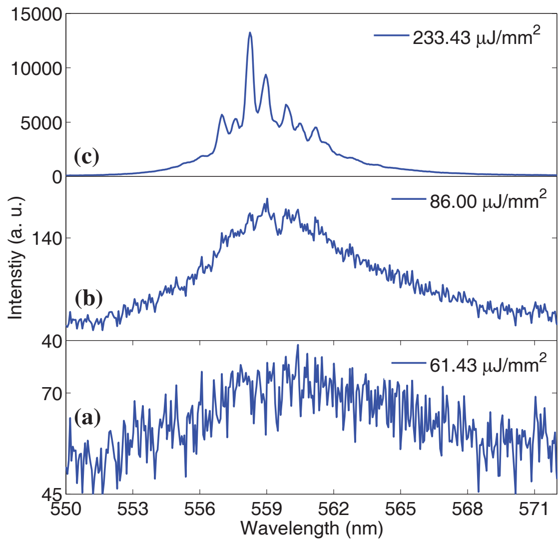

Fig. 15. (Color online) SPECTRAL BEHAVIOR OF MICROFLUIDIC RANDOM LASER: Emission from the MFC with increasing pump powers is shown. (a) Initially, at low pump fluence below the lasing threshold, a single broad spontaneous emission peak is observed. (b) The emission spectrum narrows as the pump fluence increases close to the lasing threshold ($\sim$ 80 μJ/mm^2). (c) At fluences beyond the lasing threshold, very narrow peaks at random spectral positions are observed, i.e., random lasing with coherent feedback.

Random laser action is illustrated in Fig. 15. At low fluence the spectrum consists of a single broad spontaneous emission peak which narrows when the pump fluence increases. The narrowing of the emission bandwidth with the increase in pump power is observed in Figs. 15(a) and 15(b). This is characteristic of ASE. Above the threshold found to be about 80 μJ/mm^2, very narrow randomly positioned peaks appear in the emission spectrum as seen in Fig. 15(c). The measured linewidth on the order of 0.3 nm is instrument limited. These lasing peaks are the signature of a coherent feedback mechanism. The feedback is provided by multiple scattering along the channel.

Spectral sensitivity of the laser emission on the pumped region is then investigated by changing the pump stripe position and length by scanning a slit with adjustable aperture in front of the sample. Verification is provided by monitoring the spectral peaks as the position of the pump stripe is changed. This method is quite similar to that used by van der Molen *et al.*,[50] discussed in Sec. 2.3. The spatial and spectral dependence of the laser emission is shown in Figs. 16(a) and 16(b) for two different pump stripe lengths 300 μm and 800 μm, respectively. Stochastic emission dependence on local pumping is observed; different modes appear in the emission spectrum when the spatial pumping conditions are varied. Different nonoverlapping pumped areas correspond to different independent random configurations.[80] Thus, several unique lasers can be generated within a single microfluidic random laser.

These experimental results are supported by 1D simulations based on the transfer matrix method (see Sec. 4.1). Gain and absorption are simulated using a complex index of refraction assumed to be frequency independent. Because we are mostly interested in behavior near threshold, the gain is assumed to be linear in the sense that it does not depend on the field intensity. The location and size of the pumped region, the absorption length, and the system size are adjusted to be comparable to experimental parameters. The system is randomized by specifying different thicknesses for the polymer layers with $\langle d_1 \rangle = 6$ μm and $\eta = 0.17$. The lasing frequencies and thresholds for each mode satisfying outgoing-only boundary conditions are found within the experimental wavelength range shown in Fig. 16. The localization length of the simulated structure is found to be $\xi = 44$ mm, which is much larger than the system size $L = 3$ mm. As this time-independent numerical method is only valid at or below threshold,[84] the spectrum is artificially reconstructed. Assuming the lower threshold modes will appear with greater strength in the emission spectra, each mode is given a spectral intensity proportional to the inverse square of its threshold. The position of the pump is moved across the length of the structure like in the experiment. The results of these simulations shown in Fig. 17 are in good qualitative agreement with the experiment. As in Fig 16, different modes appear in the spectrum when the spatial pumping conditions are varied. The peak position changes with a spatial correlation length, which is dependent on the pump stripe-length. The peaks appearing in Figs. 16(a) and 16(b), for pump stripe lengths of 300 μm and 800 μm, have an average spatial correlation length

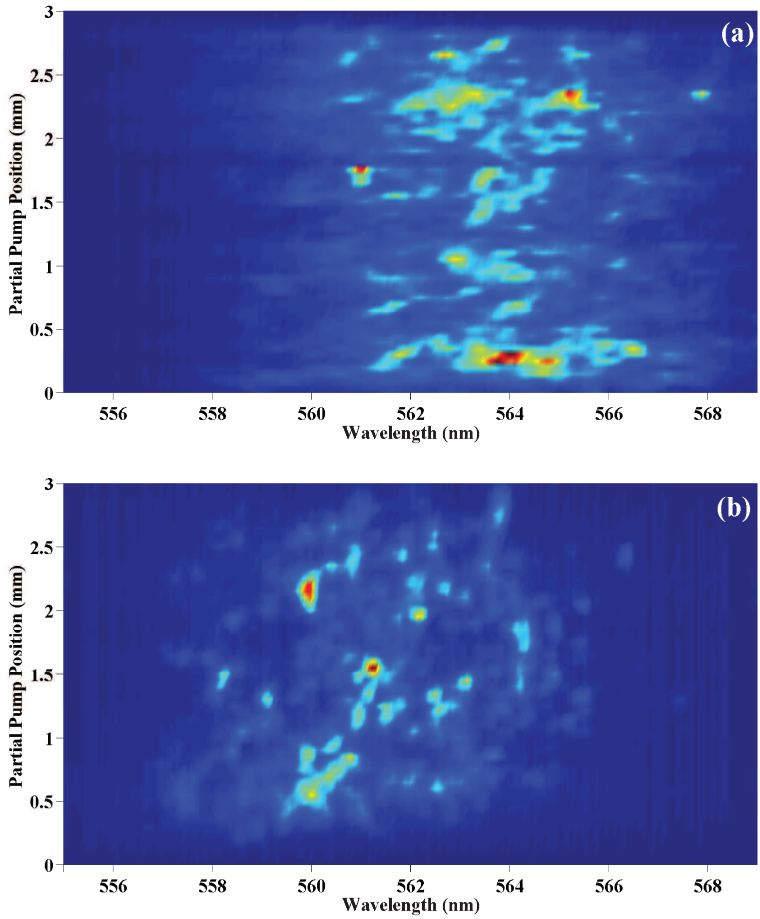

Fig. 16. (Color online) SPATIO-SPECTRAL BEHAVIOR OF MICROFLUIDIC RANDOM LASER: The partial pump stripe is translated along the length of the channel to study the spectral sensitivity of the random lasing modes to the pumped region for two different pump stripe lengths: (a) 300 μm and (b) 800 μm. The vertical axis represents the position of the partial pump stripe with respect to one end of the 3 mm long MFC. Stochastic emission dependence on local pumping is observed.

of 100 μm and 250 μm, respectively. In Fig. 17, stochastic emission dependence on local pumping can be observed for a pump stripe length of 800 μm. The peak position changes with a spatial correlation length of about 300 μm, which is in good agreement with the experimental data of Fig. 16(b).

Emission patterns along the MFC are observed and compared to simulations with good qualitative agreement. Unfortunately, a thorough examination of mode spatial properties is difficult. With weak scattering, extended modes tend to overlap spatially and spectrally, making individual mode measurement problematic. Also, as verified through numerical calculations, extended modes tend to exhibit global

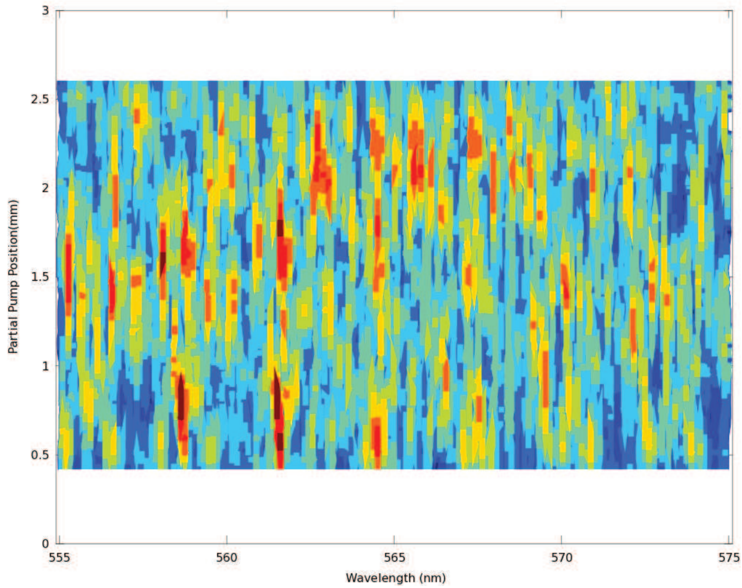

Fig. 17. (Color online) SPATIO-SPECTRAL BEHAVIOR OF NUMERICAL MICROFLUIDIC RANDOM LASER: An effective numerical spatio-spectral mapping of modes of the 1D partially pumped random laser. Here, each mode solution found is given a spectral intensity proportional to the inverse square of its calculated threshold. The partial pump stripe, of length 800 μm, is translated along the length of the random system. The vertical axis represents the position of the partial pump stripe with respect to one end of the 3 mm long system. Stochastic emission dependence on local pumping can be observed, similar to the result shown in Fig. 16(b).

intensity minima within the random system and peaks near the edges of the random system or the pump spot, whichever comes first (see Sec. 4.4). This method, therefore, does not give a precise spatial map of all extended lasing modes, since pumping at one edge may force them to disappear from the spectrum. Furthermore, whether localized or extended, mode sizes can be distorted by the size of the pump itself. Another issue is that mode coupling occurs in the multimode regime,[29] further obscuring the characteristics of individual modes. Alternate methods of collecting such spatial information are currently under investigation.

5. Toward Random Laser Control

Random lasers have raised strong interest due in large part to their simple construction, avoiding the need to engineer an optical cavity. They could therefore be a cheap alternative to conventional lasers, at least in particular cases (e.g., UV lasers[96]). The price to pay for the simplicity of the concept is that random lasers are unpredictable devices, both spectrally and spatially. Indeed, emission spectra are usually multimode, laser emission poly-directional, and lasing modes sensitive to the complete setup configuration.[3,28,29] This limits their range of applications, but does not render them useless. For example, they have found employment in speckle free imaging,[97] sensing,[94] medical diagnostics,[98] and many more. Nevertheless, for more far-reaching applications, controlling random lasers is highly desirable. The challenge, of course, lies in retaining the simple construction for applications that usually require lasers operating in a nonstochastic regime with well-controlled characteristics.

It is worth noting that in most cases described in the literature, random lasers operate in the weakly scattering regime. With the exception of a few particular examples,[99,100] the localization length is usually much larger than the sample size. Therefore the modes are spatially extended and occupy the whole system.[3,70,93,101,102] As a result, spatial mode overlap is significant in random lasers leading to complex nonlinear behavior with little control over the operation in the multimode regime. Lasing modes can strongly couple, leading to temporal oscillations[51,103] or spatial hole burning.[42] Moreover, the density of modes, in general, is high in random lasers and the lasing modes have similar thresholds.[3,93] Last but not the least, experimental and theoretical studies[36,104–107] have demonstrated that random lasers suffer from intrinsic fluctuations and instabilities, making the control of such devices even more challenging.

Despite all the difficulties, solutions have been proposed and good progress has been achieved in the direction of controlling random laser emission. The number of potential solutions has been quite large; some suggesting manipulation of the scatterers and others the gain medium or pump. Due to the topic of this review, we focus on those methods utilizing the gain as the degree of freedom. A first class of studies is aimed at reducing the range of lasing emission frequencies and restraining the direction of emission. Recently, a second class of studies based on active control of the partial pump profile were devoted to a still better control of laser emission, i.e., on-demand tuning of single-frequency emission or unidirectional emission.

5.1. *Constraining random laser emission*

In 2011, the group of Lagendijk proposed[108] the use of a dye-based random laser containing a controlled amount of nonfluorescent absorbing dye within the gain medium, in order to constrain the emission wavelength through absorption. The peak laser emission is gradually blue-shifted when more absorbing molecules are added. In the same year, Bardoux *et al.*[109] proposed a different approach based on a random distribution of InGaN/GaN quantum-disks and two-photon absorption as the pumping mechanism. Electrons decay from the two-photon excited state to an intermediate radiative state. Emission from this “virtual” state to the ground state is frequency selective. A very narrow range of emission frequencies is then observed in the resulting random laser.

In a very different scattering regime (Rayleigh), the group of Turitsyn[110] proposed a spectrally tunable random fiber laser. In a symmetric configuration, two random fiber lasers with Raman parametric amplification are connected by a band-pass filter. By adjusting the filter, laser emission is constrained and tunability is achieved over a broad wavelength range. Through direct comparisons with a conventional Raman laser, the random laser was found to exhibit a higher output power due to greater efficiency, which overcomes the larger threshold of the random laser.

In an effort to limit the number of lasing modes in a stronger scattering regime, Leonetti *et al.*[111,112] use an approach based on directional pumping, an effect first observed by Varsanyi.[44] A three-dimensional collection of nanoparticles suspended in laser dye composes the random laser. An optical pump beam illuminates not only a spot on the random sample, but two connecting triangular, or pie-shaped, spatial regions just outside the spot. Stimulated emission in the pie-shaped regions is directed along the length of the pump stripe into the random sample, which serves as an indirect but directional pump. By adjusting the pump intensity, the first modes to lase are those which are coupled to the input direction of the pump beam. Because not all modes couple to the input direction and those that do may lie anywhere within the random sample, the number of lasing modes is reduced and are generally more spatially separated from each other compared to random lasers under typical spot pumping. The ability to tune the number of excited lasing modes comes through adjustment of the maximum input angle, or the size of the pie. By increasing the allowed input angle, a broader array of modes with different coupling directions can be excited.

One effect of the ability to incrementally excite larger numbers of spatially separated lasing modes is that their overlap and interaction can be slowly adjusted. It was found that the inter-mode spectral correlation increases with the number of lasing modes as the input angle enlarges, leading eventually to a phase-locking transition.[111] Thus, according to the authors, directional pumping can be used to adjust the number of active correlated modes and to switch mode locking on and off.

A similar study, employing the directional, pie-shaped partial pumping described above, found that adjustments to the maximum input angle may also control the spatial extent of intensity within the random laser.[113] More precisely, spatio-spectral maps, which collect emitted intensity at the various lasing mode wavelengths, show that the extension of intensity increases as the maximum input angle increases. It is suspected that at larger input angles, the first lasing mode couples to other more lossy modes nearby and forces them to lase at the same frequency, thereby extending its total spatial extent. However, it was noted that a direct verification of such a mechanism is beyond current experimental abilities.

The above examples demonstrate well that some control may be wrested back from random lasers. Next, we examine a method of control that exploits some of the unpredictable properties of a random laser against itself.

5.2. *On-demand control of laser emission*

Partial pumping can be viewed as a selective pumping mechanism, which provides gain in specific spatial regions of the random laser. As seen in the localized regime in Sec. 3, this gives the pump field the ability to spatially select some lasing modes over others, similar to the naturally occurring effect of spatial hole burning.[29] In the weakly diffusive regime the situation is more complex as modes spatially overlap leading to nonlinear interaction. However, we should expect that by introducing a precisely controlled spatial modulation to the pump profile, some of the unpredictability of the random laser itself can be negated. In Sec. 4.6, partial pumping with a spot was found to separate random laser thresholds, making it easier to selectively excite one mode over the others. Combined with the facts above, properly modulating the spatial intensity distribution of the optical pump should force the laser to oscillate at a chosen wavelength.

The idea of modulating the spatial distribution of the gain was first proposed in the seminal paper on DFB lasers by Kogelnik and Shank,[114] who suggested that such a DFB structure could be designed not only by a spatial modulation of the index but also by a spatial modulation of the gain. The periodic modulation was achievable by interfering two laser beams with a variety of other methods implemented later.[115] More recently, the development of the spatial light modulator (SLM) has made more complex modulation schemes possible, with control at the micron scale of the amplitude and the phase of the light field wavefront.[116–118]

Bachelard *et al.*[119] proposed a new method to control random laser emission based on an iterative algorithm to find the optimal spatial intensity modulation of the optical pump capable of driving the random laser in the single-mode regime at a desired wavelength. They numerically demonstrated laser mode selection in a multimode random laser in the localized regime as well as in the weakly scattering regime.

A 1D random laser is modeled by a stack of 161 air and dielectric layers with respective indices n_0 and n_1. Disorder is introduced by varying randomly the thickness of the layers. The gain is described by a frequency-dependent susceptibility introduced in the dielectric layers,

$$\chi_g(k) = \frac{A_e N_{\text{pump}}}{k_a^2 - k^2 - ik\Delta k_a}\,, \tag{8}$$

where A_e is a material constant, N_{pump} the maximum density of excited atoms for uniform pumping, and k_a and Δk_a are the atomic transition and spectral linewidth, respectively. The dielectric index within the scatterers becomes

$$n_1 \to \sqrt{n_1^2 + f_e(x)\chi_g(k)}\,, \tag{9}$$

where $f_e(x) \in [0, 1]$ mimics the spatial modulation along x of the pump excitation. Using the transfer matrix method (described in Sec. 4.1), lasing modes are computed for a given pump profile f_e. To find the optimal profile that gives single-mode lasing at a given wavelength, the threshold of the targeted mode is compared to the smallest threshold of other lasing modes in the emission spectrum. The projected gradient method is applied to iteratively minimize this ratio.

To test the method, the strong scattering regime was first considered (high contrast index $\Delta n = n_1 - n_0 = 0.60$). In this regime where the localization length is smaller than the system size, modes are spatially confined [see Fig. 19(a)] and spectrally well separated. Therefore, local pumping easily selects them, as demonstrated earlier[38,39] (see Sec. 3). It is convenient to represent each lasing mode in

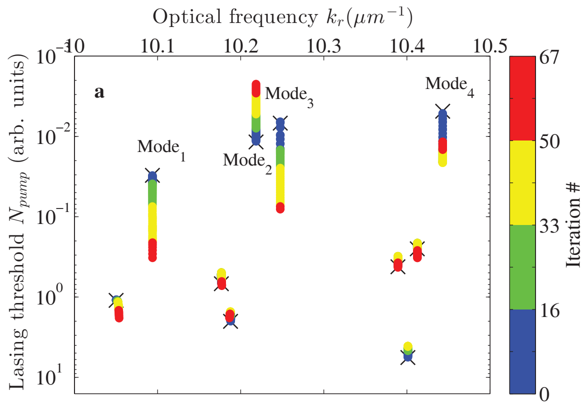

Fig. 18. (Color online) LASING MODE BEHAVIOR DURING OPTIMIZATION WITH STRONG SCATTERING: 1D Numerical optimization in the localized regime when index contrast is high ($\Delta n = 0.6$). Modes are computed using the transfer matrix method. Each mode is described by its lasing threshold N_{pump} and corresponding optical frequency k_r. Crosses: Lasing modes when the 1D random laser is uniformly pumped. In this case, Mode$_3$ would be the first to reach the lasing threshold. For Mode$_2$ to lase first instead, the iterative method is applied to find the optimized pump profile which would select Mode$_2$ at the expense of other modes. Full dots: lasing modes at successive iteration steps. After 67 iterations (red dots), the lasing threshold of Mode$_2$ has diminished while the thresholds of other competing modes increase. Hence, Mode$_2$ has been selected.

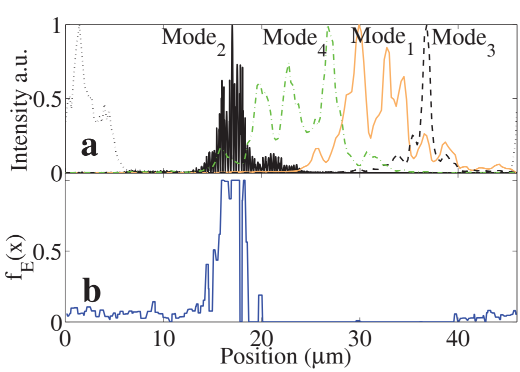

Fig. 19. (Color online) LASING MODE SELECTION WITH STRONG SCATTERING: (a) Spatial profile of the amplitude of 5 localized modes. (b) The optimized pump profile obtained after selection of Mode$_2$ presented in Fig. 18. This pump profile is optimized to locally pump Mode$_2$ and concentrate the gain distribution around this mode.

the complex plane, with its optical frequency k_r on the real axis and its threshold value $N_{\rm pump}$ on the imaginary axis as in Fig. 18. After 67 iterations, modes spectrally close to $\rm Mode_2$ see their thresholds significantly increased. Meanwhile, the threshold of $\rm Mode_2$ decreases as the gain is now optimally spatially distributed for this particular lasing mode (see Fig. 19). Therefore, $\rm Mode_2$ has been selected and would be the first to lase when the optimized pump profile is applied. This profile is found, as expected, to be spatially localized at the location of $\rm Mode_2$ [see Fig. 19(b)].

The method is next applied to a weakly scattering random laser. The weakly scattering regime is more challenging as lasing modes are spatially extended and significantly overlap [see Fig. 21(a)], making spectral selection of a given mode impossible *a priori*. The index contrast ($\Delta n = n_1 - n_0 = 0.05$) was chosen so that the localization length is much larger than the system size. The iterative selection of a particular mode is shown in Fig. 20. Surprisingly, the optimization process still works efficiently, yielding a complex spatial profile [seen in Fig. 21(b)] that optimizes pump injection in the desired mode while working to the disadvantage of all other modes. Actually, this iterative process is highly nonlinear. Because the complex susceptibility depends locally on the pump intensity, the refractive index distribution is slightly altered by the nonuniform pump profile. Bachelard *et al.* showed that, surprisingly enough, the system spontaneously self-adjusts to modify the lasing modes and favors the selected one.

This method of optimizing the pump profile is, in fact, quite general and can be used to control other characteristics of a random laser such as directivity. Very recently, the group of Rotter[120] has numerically demonstrated that a 2D random

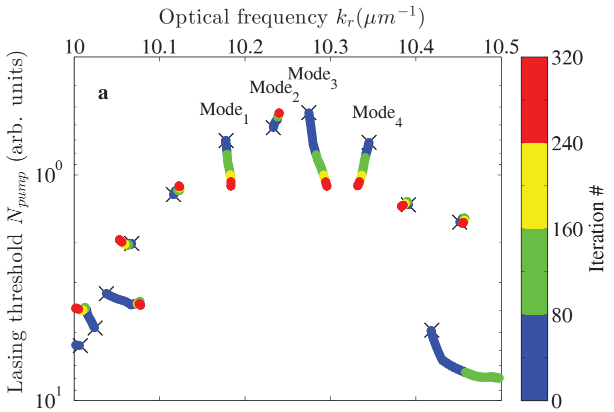

Fig. 20. (Color online) LASING MODE BEHAVIOR DURING OPTIMIZATION WITH WEAK SCATTERING: 1D Numerical optimization in the weakly scattering regime when index contrast is low ($\Delta n = 0.05$). Same as in Fig. 18, after 320 iterations (red dots), the lasing threshold of $\rm Mode_2$ has diminished slightly while other competing modes see their threshold increase. Hence, $\rm Mode_2$ has been selected.

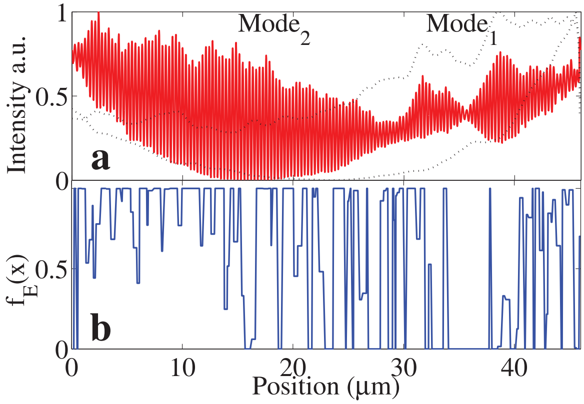

Fig. 21. (Color online) LASING MODE SELECTION WITH WEAK SCATTERING: (a) Spatial profile of the amplitude of 2 lasing modes for a uniform pump profile in the weakly scattering regime (index contrast $\Delta n = 0.05$). Modes are extended in this regime of weak scattering and strongly overlap. (b) The optimized pump profile obtained after selection of Mode$_2$.

laser can be controlled using iterative pump shaping to enforce laser emission in a given direction, within a given angular range.

The concept of pump shaping represents a breakthrough for random lasers applications. There is, *a priori*, no limitation on the geometrical shape of the pump, hence the potential for precise control. It was shown that with the right choice of the optimization algorithm, this technique is extremely robust. The method might also be extended to other classes of lasers, such as DFB or broad-area semiconductor lasers (with complex spatiotemporal dynamics[121]). Laser brightness may be optimized in such cases, or unwanted nonlinear effects, such as filamentation,[122,123] may be minimized.

6. Conclusion

Due to its complex characteristics, a random laser can display versatile behavior spatially, spectrally and temporally, in response to different excitation conditions. Thus, spatial control of the pump profile has significant consequences for the light emission of a random laser, in sharp contrast to conventional lasers made of simple cavities. Because of their randomness, changing the position, shape, and size of the pumped domain or the intensity profile of the pump amounts to exploring different system configurations. This leads to a laser field, whose main features like frequency, emission location or direction are very sensitive to the pump configuration. A striking example is pumping a small region of the random system, which reduces the effective system size when absorption outside the pumped domain is significant. The confinement of lasing modes results from an inhomogeneous gain/absorption distribution instead of strong scattering and localization. Another major consequence of partial pumping in random lasers is to modify the nature of the lasing modes. One benefit of this is that the laser output is not bound by the parameters of the passive random system (even though passive systems have proven themselves quite useful[117,118,124–130]). However, except in the 1D systems described in this review, the relation of new lasing modes to the QB states or to the lasing modes of the uniformly pumped system, is not entirely clear. Unlike most conventional lasers that have almost closed cavities, random lasers are very open, leading to significant spatial and spectral overlap of quasimodes. In the presence of gain, this results in strong mode competition. Hence, mode coupling via self- and cross-saturation effects and other nonlinear interactions are likely to be significantly modified by the pump configuration.

Because of all these distinctive features, partial pumping can be used to control random laser performance and extract diverse behavior. For example, simply changing the size of pump volume can vary the number of lasing modes and tune the spatial coherence of laser emission.[131] The low spatial coherence in combination with high radiance makes random lasers an ideal illumination source for parallel imaging and projection.[97] Very recently, as described in Sec. 5.2, employing a more complex shape to the pump field has been shown to allow on-demand tunability of random lasers. A finely controlled amount of randomness introduced to the optical pump field produces more predictable laser output. An advantage here is that neither careful engineering of an optical cavity nor manipulation of the random scatterer configuration is required. The random laser, whatever the composition of the scatterers and gain medium, is taken as is, with only the pump field requiring adjustment. The bulk of such work to date has been done numerically, revealing the possibility of choosing the lasing frequency and directionality of random lasers. However, the concept has been experimentally demonstrated in random systems with strong scattering[132] and experiments are currently underway to explore the practicality of such an approach in the more common weakly scattering case.[133] For future practical applications, electrical pumping[134,135] will provide a cheaper and more convenient pumping source.[136] Partial electrical pumping has been implemented for microcavity lasers[8–10] and found some success in the excitation of specific mode types. However, the electrode patterns are shaped prior to the experiment, making the flexibility of electrical pumping perhaps an issue to address in random lasers.

In addition to the great potential that partial pumping has for random laser control, it also raises many fundamental questions that need to be answered like the nature of the lasing modes, which depend on the interplay between the material, structure of the random laser, and the pump configuration. This involves numerous aspects, such as the dependence of partial pumping effects on the degree of scattering and disorder of the random system inside and outside the pumped domain. Contrary to the case of uniform pumping, a complete theory of partial pumping remains to be developed. Further studies on partial pumping are needed for a deep understanding of the underlying physics and for the control of random laser emission which is a major issue for future applications.

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