Selective Excitation of Localized Modes in Active Random Media

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Laser action in the regime of Anderson localization is investigated by coupling Maxwell's equations in a strongly scattering medium with the rate equations of a four-level atomic system. We find that the localized modes of the passive medium are not modified by the presence of gain and serve as resonant microcavities of the random laser. The spectrum of laser emission is shown to depend on the location of the external pump. These results are similar to the recent experimental observations in semiconductor powder [Cao *et al.*, Phys. Rev. Lett. **82**, 2278 (1999)]. We show that local pumping of the system allows selective excitation of individual localized modes.

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After the pioneer work of Lethokov in 1968 [1], laser action in mirrorless scattering media has recently received considerable interest. If this active field of research is motivated by the possibility of future applications [2], it also raises fundamental interrogations. Among them, how laser action depends on the strength of the disorder and, for highly scattering media, how laser gain affects Anderson localization are still open issues.

Anderson localization of waves, originally predicted in the context of electronic transport [3] and extended later to electromagnetic waves [4], is often described in terms of transport properties. As a result of strong interferences and multiple scattering, a wave packet may have a finite probability of return to the same site, which results in a vanishing diffusion coefficient in an infinite medium and an exponential decay of transmission, characterized by the average localization length, ξ. These transport properties directly originate from the confined structure of the eigenmodes. The localized modes can be seen as complex microcavities, described asymptotically by an exponential decay of their envelope on a typical length ξ.

One interesting issue when considering single localized eigenmodes of a specific realization of the disorder is the possible role these microcavities could play in the presence of gain. Numerical simulations of random active media using a phenomenological description of the gain have indeed suggested the principle of a "distributed feedback random cavity laser," where the localized modes of the passive medium could serve as the feedback cavity of the laser [5]. The first reports of laser action in random media [6–8] showed, however, no evidence of resonant feedback due to localized modes. The linewidth narrowing of the emission spectrum was interpreted in terms of nonresonant feedback of spontaneous emission amplified along open scattering paths [9–12].

Only recently has laser action in random media with resonant feedback been reported [13,14]. Cao *et al.* used a semiconductor powder, which played simultaneously the role of the random and the active medium. Above a certain threshold, sharp peaks emerged in the emission spectrum. This behavior has been identified as a direct manifestation of the resonant feedback process provided collectively by the strongly scattering medium. Here, closed loop paths formed by the scatterers serve as laser cavities. Because these cavities can be spatially selected, different peaks appeared for different locations of the pump excitation. If evidence of strong scattering regime has been brought, possible connection with Anderson localization was merely suggested [15]. Moreover, no complete theory of strong localization for active random media is yet available. One may wonder, for instance, whether the localized modes of the passive random system are preserved or, on the contrary, to which extent they are modified by the gain. One may even ask further if localization is enhanced or inhibited in the presence of gain [16–18].

In this Letter, we examine the role of strong localization in the lasing action process. We use a numerical model which describes the full dynamics of the field and the levels' populations in two-dimensional (2D) active random media. First, we select a window of modes strongly localized in the spectrum of the passive medium and examine the spatial and spectral characteristics of these modes. Next, the passive modes are compared with the laser modes when the gain is activated. One of the main results presented here is that the active medium is described by the modes of the passive system. Aside from a small frequency pulling effect due to the amplifying medium, the laser frequencies coincide with the eigenfrequencies of the passive system and the 2D spatial profile of the localized wave functions are remarkably well reproduced. Neither enhancement nor destruction of strong localization are observed. Next, we demonstrate how lasing frequency control via the selective excitation of individual localized modes can be achieved by local pumping of the system.

We consider a two-dimensional disordered medium of size L^2 made of circular scatterers with radius r, optical index n_2, and surface filling fraction ϕ, imbedded in a matrix of index n_1. This system is equivalent to a random array of dielectric cylinders oriented along the z direction. The matrix is chosen as the active part of the medium, in order

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to control the randomness and the gain independently. Following the conventional laser equations [19], we describe the dynamics of atomic populations by rate equations of a four level atomic system and the time evolution of the field by Maxwell's equations, including a polarization term due to atomic population inversion. The corresponding equations are identical to those laid out in [20] where key aspects of the experimental observations of Cao *et al.* have been successfully reproduced in one-dimensional systems. Here, we consider the E_z, H_x, and H_y components of a 2D TM field. The FDTD (finite-difference time-domain) method [21] has been used to solve the Maxwell's equations. We use PML (perfectly matched layer) absorbing boundary conditions [22] in order to model an open system. A time step of 2.10^{-17} s has been chosen to describe the time evolution of the optical field ($\nu^{-1} \sim 10^{-15}$ s). Although atomic parameters of dye molecules have been preferred for their relatively short emission lifetime (10^{-9}–10^{-10} s), the dynamic range of times spans 7 to 8 orders of magnitude. However, in the calculations presented here, the stationary regimes are reached after 10^5 to 10^6 time steps, because of the relatively short decay times of the modes of our systems (see below).

We first study the modes of the passive system (without gain). We choose $L = 5.5$ μm, $n_1 = 1$, $n_2 = 2$, $r = 60$ nm, and $\phi = 40\%$ (see Fig. 4). The time response to a short pulse is recorded and Fourier transformed to get the power spectrum. Figure 1a shows two spectra corresponding to the Fourier transform of the first and second half of the time record of the field. The two spectra are different because the system is open and supports only leaky modes or "quasimodes" [23]. The most leaky modes observed in the first spectrum are strongly attenuated in the second spectrum where only modes with the longest lifetimes still survive. We can excite these modes separately by a monochromatic source at the eigenfrequencies measured in the spectrum, to characterize their spatial profile and time evolution. Figures 2a and 2b show two examples of eigenstates, called a and b, obtained following such a resonant excitation. They correspond to $\lambda = 446.9$ and $\lambda = 445.0$ nm in the spectrum of Fig. 1a. Besides their complex geometries and random locations in the system, these modes are characterized by a strong spatial localization. A semilog plot would show an exponentially decaying envelop. The measured characteristic decay length is close to 0.5 μm, which is about ten times smaller than the system size $L = 5.5$ μm. Accurate measurement of the decay time of the modes gives $\tau = 3.49$ and 0.72 ps for modes a and b, respectively, corresponding to a quality factor $\nu/\delta\nu$ equal to 14 700 and 3030, where $\delta\nu$ is the linewidth. Even for mode b, leakage is weak enough for this mode to survive over times of the order of 10^{-12} s. A rough estimate of the mean level spacing $\Delta\nu$ gives a Thouless number $\delta\nu/\Delta\nu$ smaller than unity [24]. The localized nature of the modes and their spectral properties confirm

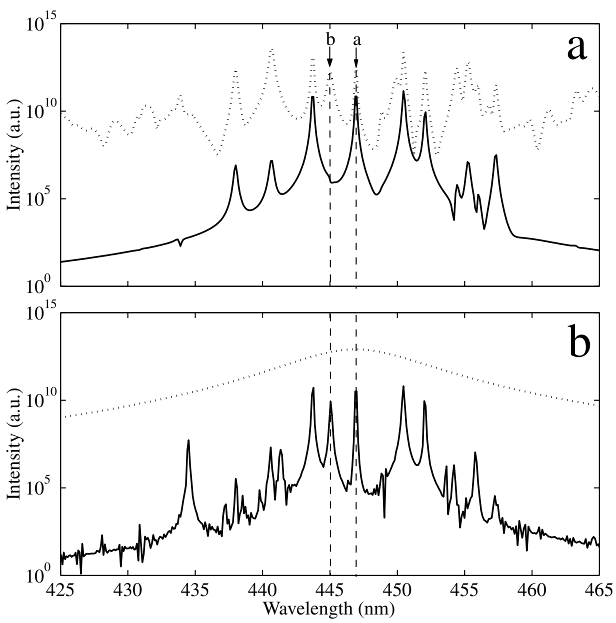

FIG. 1. (a) Semilog representation of the power spectra corresponding to the Fourier transform of the first (dotted line) and second (full line) half of the time record of the field. (b) Semilog representation of the power spectrum of the laser field at high pumping rate (full line). The (rescaled) gain profile is represented in dotted line.

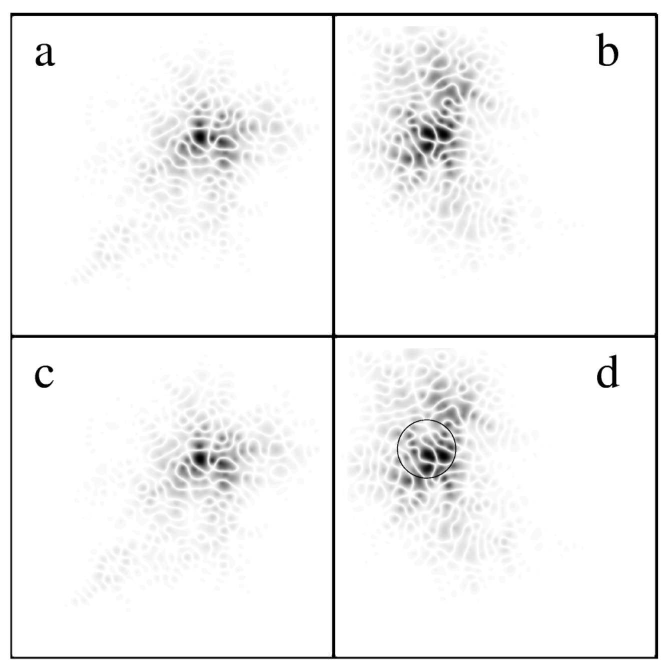

FIG. 2. Spatial distribution of the field magnitude of (a) mode a ($\lambda = 446.9$ nm) and (b) mode b ($\lambda = 445.0$ nm) of the passive system. Spatial representation of the magnitude of the laser field just above threshold for extended (c) and spatially localized (d) gain. Note that amplitudes in (a) and (c) or (b) and (d) are not comparable since they have been scaled differently to use the full color map. The circle represents the spatial extension at σ of the Gaussian pump.

that the regime of Anderson localization has been reached within the limited dimensions of the system.

In a second step, gain is introduced in the system by pumping the four level atoms uniformly over the whole system. The maximum of the gain curve is positioned at $\lambda = 446.9$ nm (at exact resonance frequency of mode a). The small background field that is necessary to initiate lasing action is provided by a low intensity broadband seed pulse. Just above threshold, a stationary regime is reached after a transient exponential growth of the field amplitude. The corresponding map is displayed in Fig. 2c. One recognizes the pattern of mode a of the passive system (Fig. 2a). We deliberately chose to represent the amplitude of the field rather than the intensity for a better display of the tails of the modes. A more quantitative comparison of the normalized intensities of the passive mode and the laser mode (averaged over one thousand periods) reveals a maximum relative difference of 4.7% for local intensities of the mode as low as 1% of its maximum value. The corresponding spectrum (not shown) exhibits a unique peak at $\lambda = 446.9$ nm. This result demonstrates clearly that the localized mode is responsible for the frequency selection of the random laser and that the introduction of gain in the disordered system does not modify the structure of this mode.

At higher pump levels, the laser emission is multimode. After a transient regime lasting a few picoseconds, the field becomes stationary on average though exhibiting beats between several excited modes. The field map at a particular time is shown in Fig. 3. One clearly recognizes a mixture of patterns including the two modes a and b of Fig. 2. This shows that the laser field corresponds to a superposition of modes of the passive system. This result is confirmed by the spectrum of the laser field shown in Fig. 1b. The narrow peaks correspond to the eigenfrequencies of the passive system, aside from a small frequency pulling effect (relative frequency shift less than 5×10^{-4}).

If mode a, which has the longest lifetime in the frequency range considered, can be excited alone by adjusting the pumping rate near threshold, other modes may also be selected individually. One possibility would be to shift the maximum of the gain curve close to another eigenfrequency of the passive system. Though realizable in numerical calculations, the gain curve is fixed by the choice of the active material and cannot be adjusted in actual experiments. Another possibility, which is used for the selection of individual modes of a classical laser, is to introduce an etalon into the cavity. For obvious reasons, this method is also not possible for a random laser. We show now that it is possible to take advantage of the nature of the localized modes to select them individually. For this purpose the system is not pumped uniformly but locally. We choose an external pump with a Gaussian spatial profile of width of the order of ξ, $\sigma \approx 0.5$ μm. Since the localized modes have a complex geometry and have different locations in the system, the overlap of the gain with a given localized mode can be maximized by adjusting the position of the pump. By scanning the system with the pump, we have been able to selectively excite each of the modes corresponding to the highest peaks of Fig. 1a. Local excitation of the system is illustrated in Fig. 4, which displays the spatial distribution of the atomic population difference ΔN between the upper and lower lasing levels, in the

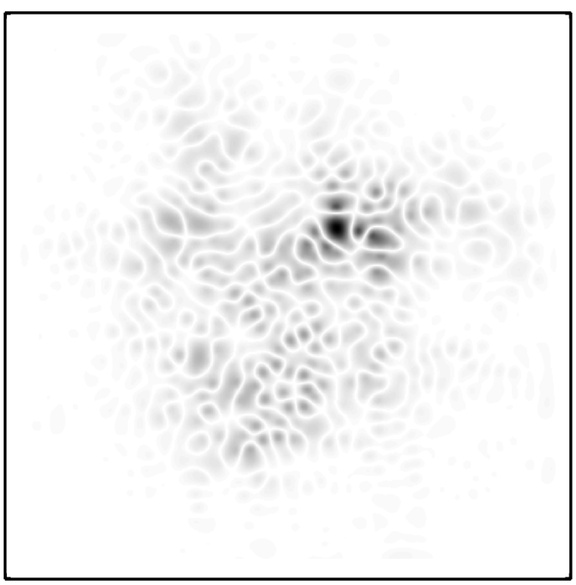

FIG. 3. Spatial distribution of the magnitude of the laser field at a given time, at high pumping rate. Corresponding spectrum is shown in Fig. 1b.

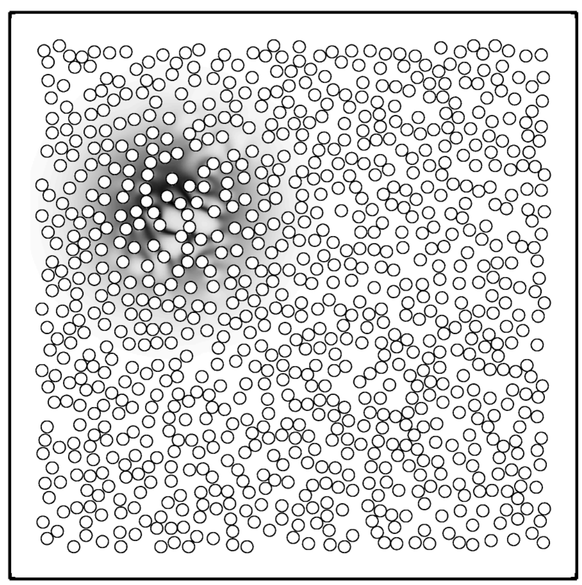

FIG. 4. Gray scale representation of the spatial distribution of the atomic population difference ΔN between the upper and lower lasing levels for a local excitation of the system by a Gaussian pump of width $\sigma = 0.5$ μm. The white spots inside the excited region indicate the gain saturation. The small circles represent the scatterers.

stationary regime. Population inversion at the position of the external pump is seen. Saturation of the gain is also observed, as revealed by the spots where ΔN is strongly reduced inside the active region. This is the equivalent for the random laser of the hole burning effect, well known in conventional lasers [19,23]. Figure 2d shows the map of the corresponding laser mode. One recognizes mode b of the passive system (Fig. 2b). Although farther from the maximum of the gain curve ($\lambda = 446.9$ nm) and more dissipative than mode a, this mode has been excited alone, as confirmed by the single peak located at $\lambda = 445.0$ nm in the corresponding power spectrum (not shown). It is interesting to note that the overlap of the pump profile and the excited mode does not need to be perfect. In fact, it is sufficient for the difference between the gain and the losses to be maximum for this mode at the position of the external pump. Thus, this example demonstrates that it is possible to force the random laser to oscillate in a single mode different from the most favorable one. As expected, the pumping rate must be adjusted for each new position of the pump in order to excite a single mode. In other words, the local threshold for lasing depends on the position of the pump. Finally, it is interesting to consider the case where the external pump is located near the boundaries of the system. Since leakage is important, the modes positioned at those locations are strongly damped. However, by using a high pumping rate, we have been able to excite such modes. This shows that lasing action is not limited to the best modes of the system. This result is relevant for actual experiments since external pumping of a strongly scattering system is likely to be more efficient at its boundaries.

In conclusion, we have demonstrated numerically by coupling the Maxwell's equations in a disordered medium with the rate equations of a four level atomic system that the lasing modes are identical to the passive modes without gain. When the external pump is focused, the lasing modes change with the location of the pump. This result is in agreement with experimental observations reported by Cao *et al.* [13]. By properly adjusting the pumping rate, it is possible to excite individually different modes of the passive system. These results show that introducing gain could help to study wave propagation in strongly scattering systems. An interesting question would be to know, for instance, if the introduction of gain would make it possible to discriminate between the diffusive and localized regimes, which remains a challenge in actual experiments.

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