Lasing with Resonant Feedback in Weakly Scattering Random Systems

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Laser action in active random media in the weak scattering regime far from Anderson localization is investigated by coupling Maxwell's equations with the rate equations of a four-level atomic system. We report systematic lasing action with resonant feedback and show that the lasing modes mostly consist of traveling waves spatially extended over the whole system. Next we address the question of the origin of the feedback mechanism in such a system where no disorder-induced long-lived resonances are available, and present strong evidence that they correspond to the quasimodes of the passive system. This in turn provides an original way to access the spatial distribution of the quasimodes of a non-Hermitian system.

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Lasing with intensity feedback and lasing with field feedback are the two acknowledged types of random laser emission [1]. As first predicted by Lethokhov [2], lasing with intensity (nonresonant) feedback (LIF) is based on photon diffusion with energy feedback only. In a disordered open system, the distinct effect of the scatterers is to increase the length of the light paths before photons escape from the system. Hence, amplification is improved by randomness as compared to an homogeneous active medium. Like conventional amplified spontaneous emission (ASE), there is a threshold above which the light emission increases rapidly with the pumping rate and the linewidth narrows abruptly down to values of a few nanometers. Lasing with field (resonant) feedback (LFF) is similar to lasing in a conventional cavity. The feedback due to randomly distributed scatterers leads to the coherent oscillation of a radiation mode where not only amplitude but also phase conditions are satisfied [3]. These modes show themselves plainly above some threshold of the pumping rate like discrete spectral lines as narrow as one-tenth of a nanometer, which appear on top of the broad fluorescence band.

Historically, LIF was first observed in moderately scattering systems in the diffusive regime [1]. More recently, LFF has been observed in semiconductor powders, conjugated polymer films, and dye-infiltrated opals [1]. These last experimental observations have been tentatively explained in terms of confined modes such as localized modes [4]. Indeed, random lasing with resonant feedback is naturally expected if light is localized [5,6]. However, among the numerous experimental systems that have exhibited LFF, many of them seem to be too far from the Anderson localization regime to be able to support confined modes (see, for instance, [7]). Hence, the nature of the lasing modes in such systems is an open question. Several mechanisms have been proposed to explain LFF in diffusive random systems in terms of anomalously localized modes [8], or the absorption-induced localization of lasing modes in the local pump region [9], or the amplification of spontaneous emission along very long paths [7]. Although they are possibly achieved in some specific situations, such mechanisms cannot explain the whole set of experimental observations of LFF.

In this Letter, we study active random media in the weak-scattering regime. We present numerical simulations which demonstrate lasing action with resonant feedback, despite the field feedback is very weak in the systems we consider. Lasing action is observed for any weakly scattering system, independently of its parameters and disorder realization, inasmuch as the pumping rate is large enough. In contrast to the lasing modes in the localized regime, here the lasing modes mostly consist of traveling waves spatially extended over the whole system. We provide strong evidence that the first lasing modes are the short-lived quasimodes of the passive system. This result suggests an original method to obtain the spatial distribution of electromagnetic quasimodes of an open system even in the presence of strong leakage.

We consider a two-dimensional (2D) system of size $L^2 = 5 \times 5\ \mu\text{m}^2$ made of circular particles with radius $r = 60$ nm, optical index $n_2 = 1.25$, and surface filling fraction $\Phi = 40\%$, which are randomly distributed in a background medium of index $n_1 = 1$ (inset of Fig. 1). This system can be considered as a random collection of cylinders oriented along the z axis. The background medium is chosen as the active part of the system and is modeled as a four-level atomic system. We describe the time evolution of the atomic populations by rate equations and the time evolution of the field by the Maxwell's equations including a polarization term due to the atomic population inversion. We have used perfectly matched layer (PML) absorbing conditions in order to model an open system [10]. The corresponding equations are identical to those that have been used in [6]. The value $n_2 = 1.25$ is the only but crucial difference with the 2D system studied in [6] where the particle optical index n_2 was chosen equal to 2. It was

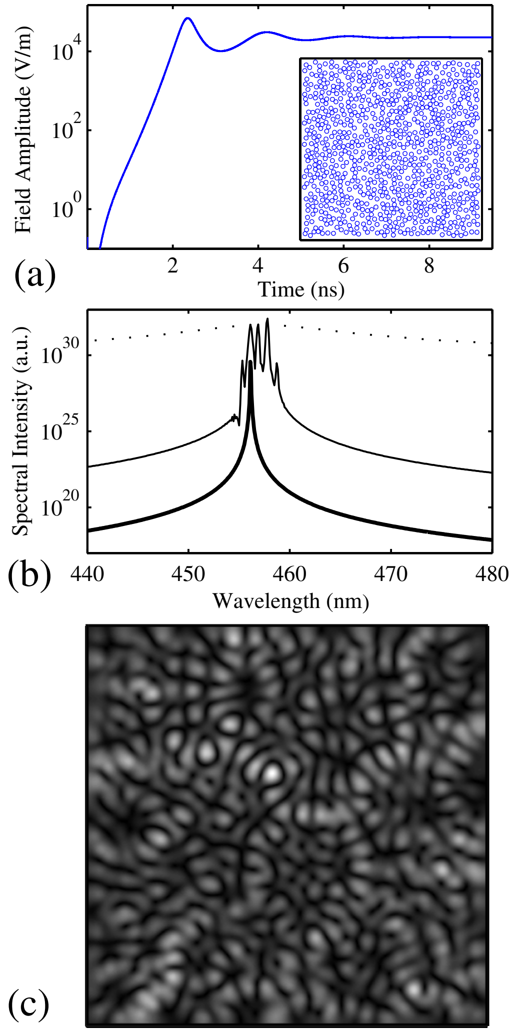

FIG. 1 (color online). For the spatial arrangement of the scatterers shown in the inset: (a) Dynamics of the field amplitude. (b) Emission spectrum in the steady state regime with one mode (thick line) and with several modes at higher pumping rate (thin line) compared to the gain profile (dotted line). (c) Spatial distribution of the field amplitude.

shown in [6] that for $n_2 = 2$, passive systems exhibit high-quality localized modes. When gain is added to such systems, lasing modes are found identical to the modes of the passive system demonstrating that the modes of a random laser in the Anderson localization regime behave as the cavity modes of a conventional laser. For $n_2 = 1.25$, we have investigated several systems corresponding to different sizes and different realizations of the disorder. The mean free path has been estimated to be not much smaller than L. This weakly scattering regime, which is intermediate between ballistic and diffusive, is thus far from Anderson localization. In this regime that we shall simply call diffusive, all the energy initially put inside the system flows out rapidly through the open boundaries and no mode survives long enough to be spectrally identified from the impulse response.

Despite this weakly scattering regime and the absence of identifiable resonances in these random systems, lasing action with resonant feedback is demonstrated. The pumping rate is adjusted just above threshold. After an exponential growth of the intensity, strong relaxation oscillations which are characteristic of a damped laser cavity are observed [Fig. 1(a)]. The transient dynamics is very similar to what was observed in localized random active media, except that the pump level can be several orders of magnitude higher in weakly scattering media in order to achieve single-mode lasing. Eventually, the steady state is reached, associated with a sharp single peak in the emission spectrum [thick line in Fig. 1(b)] with a linewidth only limited by the finite time record length of the simulation. At higher pumping, additional lasing peaks appear [thin line in Fig. 1(b)]. Like in the localized case [6], the dynamics and the discrete lasing frequencies are hallmarks of lasing action with resonant feedback. However, the lasing modes in both regimes present differences. First, the lasing mode in the localized system is spatially confined [e.g., Fig. (2c) in [6]] following closely the distribution of a localized mode, while in the diffusive sample, the lasing mode is distributed over the whole system, as shown in Fig. 1(c). Next, the lasing mode is a standing wave in the localized regime while it has a large traveling component [11] in the diffusive regime; i.e., the field is "complex" [12]. A general expression for the electric field of the single lasing mode is, $E(\vec{r}, t) = A(\vec{r}) \cos[\omega t + \phi(\vec{r})]$, where $A(\vec{r})$, and $\phi(\vec{r})$ are, respectively, the amplitude and the phase of the field. For the localized lasing mode, $\phi(\vec{r})$ takes only two values, $\phi(\vec{r}) = \phi_0$ and $\phi_0 + \pi$, where ϕ_0 is arbitrary, which means that the mode is a standing wave. This is illustrated in Fig. 2(a) by the probability distribution of the phase, $\phi(\vec{r})$, which shows two peaks at ϕ_0 and $\phi_0 + \pi$ in the localized regime. In the diffusive regime, however, the phase spreads over the full range $[0, 2\pi]$ [13]; i.e., the traveling wave component dominates.

It is convenient to introduce the spatial correlation function $C_{\mathcal{E}}(t_0, t) = \iint d^2\vec{r}\mathcal{E}(\vec{r}, t_0)\mathcal{E}(\vec{r}, t)$ of the normalized field $\mathcal{E}(\vec{r}, t) = E(\vec{r}, t)/[\iint d^2\vec{r}E^2(\vec{r}, t)]^{1/2}$. For a purely standing wave, the normalized field map $\mathcal{E}(\vec{r}, t)$, should not change although the field $E(\vec{r}, t)$ oscillates in time. Therefore, the correlation function versus time is expected to be a square wave function jumping every half-period between $+1$ (in-phase) and -1 (180° out of phase). This is what is observed for lasing modes in the localized regime [Fig. 2(b)]. Instead, in the diffusive regime, the correlation function exhibits a sinuslike oscillation between $+1$ and -1. This indicates that the normalized field map is continuously changing over a period and is recovered every half-period with opposite sign [14,15].

The question remains of the origin of the resonant feedback mechanism in the diffusive regime. To answer this question, it would have been natural to compare the lasing modes with the modes of the passive cavity, as was done in

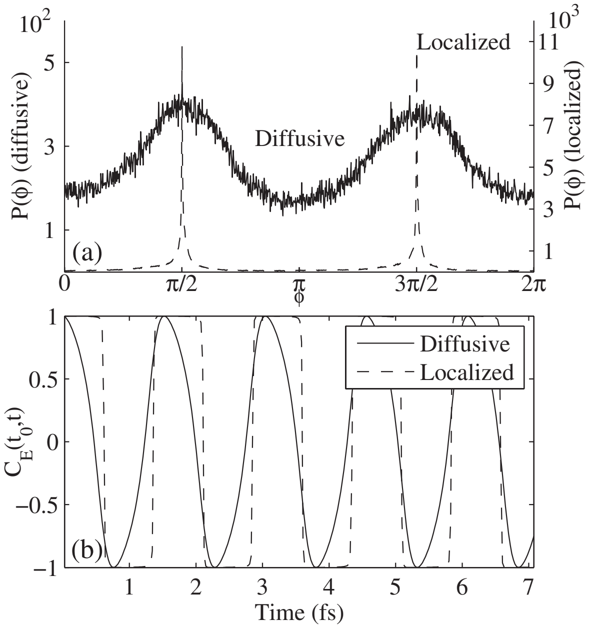

FIG. 2. (a) Probability distribution of the phase and (b) spatial correlation function of the field $C_{\mathcal{E}}(t_0, t)$ for a lasing mode in the localized (dashed line) and in the diffusive (full line) regime.

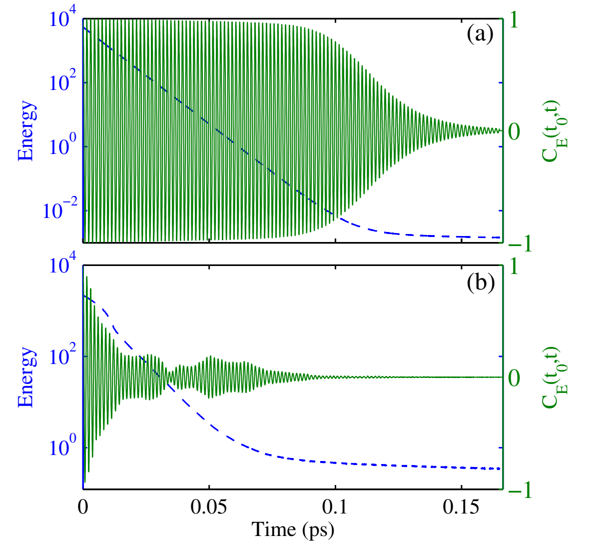

FIG. 3 (color online). Spatial correlation function of the field $C_{\mathcal{E}}(t_0, t)$ (full line) and total energy of the system (dashed line) as a function of time when excitation of the system is turned off. The initial field is (a) the laser field and (b) the field created by an arbitrary set of monochromatic sources at the laser frequency.

the localized regime, where the eigenmodes of the passive system were identified as the resonant structures of the lasing process. There, the eigenfrequencies of long-lived modes of the passive system were clearly identified by well separated peaks in the spectrum obtained by Fourier transforming the impulse response. The spatial distribution of a mode was then obtained using a monochromatic excitation at its corresponding eigenfrequency [6]. In contrast with localized states, the short-lived modes of a weakly scattering system do not yield well-defined resonances in the impulse spectrum.

Although we lack a direct comparison between the passive and lasing modes, we demonstrate indirectly that the lasing modes are indeed built on the modes of the passive system. For this purpose, the pumping is turned off at time t_0 after the lasing mode has been established and we let the field evolve by itself. Because of leakage, the field decays rapidly. The decay of the total energy of the system as a function of time is shown as the dashed line in Fig. 3(a). Damping is observed over 6 orders of magnitude and the corresponding quality factor estimated over a time interval of 0.07 ps is $Q = 30$, as compared to Q of the order of 10^4 in [6]. However, we found that its spatial distribution is reproduced identically every period, as expected for a quasimode of a non-Hermitian system. This is demonstrated in Fig. 3(a), which shows the spatial correlation function, $C_{\mathcal{E}}(t_0, t)$, of the field at time t with field at time t_0. Indeed, although the energy inside the system has decreased by several orders of magnitude, the oscillation amplitude of the correlation function remains close to 1, as long as the field amplitude has not reached the noise level. This suggests that a single short-lived resonant mode of the passive system has been excited. The field reads $E(\vec{r}, t) = A(\vec{r})\cos[\omega t + \phi(\vec{r})]\exp(-\Gamma t)$, with Γ the leakage rate, describing a "natural mode" or "quasimode", which generalizes the concept of mode to leaky systems [16]. This quasimode has the same oscillating pattern and frequency as the original lasing mode, supporting the hypothesis that lasing occurs on a mode of the passive system, even for weakly scattering systems, the only difference being its fast decay.

To validate further this result, the system in absence of gain is excited in the scattering region by several sources oscillating at the frequency of the laser. First, the amplitude and the phase of the sources are chosen constant. After the steady state is reached, the sources are turned off. We observe the energy decay at the same rate as in the previous experiment, corresponding to the diffusive escape time. However, decorrelation of the spatial distribution of the field occurs on a much shorter time scale, as shown in Fig. 3(b).

Next, sources $S(\vec{r}, t)$ are placed over the whole system and adjusted in such a way that their amplitudes and relative phases reproduce the field distribution of the lasing mode, i.e., $S(\vec{r}, t) = A(\vec{r})\cos[\omega t + \phi(\vec{r})]$. After the sources are turned off, the field decays and the result of Fig. 3(a) is recovered: the oscillations amplitude of the correlation function stays close to 1 as long as the amplitude remains above the noise level. This demonstrates that the field distribution created by this set of sources is different from the field distribution created by an arbitrary set of sources at the same frequency. Only the former describes a quasimode with $E(\vec{r}, t) = A(\vec{r})\cos[\omega t + \phi(\vec{r})]\exp(-\Gamma t)$.

Most remarkably, we were able to exhibit the same quasimode using a uniform excitation of constant amplitude A_0 with only the phase spatial distribution of the lasing mode, i.e., $S(\vec{r}, t) = A_0 \cos[\omega t + \phi(\vec{r})]$. An interference pattern builds up, which reproduces the field distribution of the quasimode. In contrast, using a source $S(\vec{r}, t) = A(\vec{r}) \cos[\omega t + \phi_0]$ with only the amplitude distribution of the lasing mode does not reproduce the field distribution of the quasimode. This result emphasizes the crucial role of the phase as expected if the laser mode is a resonance of the passive system.

Identification of quasimodes of an open diffusive system in terms of their complex eigenvalues [17] and their spatial field distribution is usually a challenging task. Here, the lasing modes provide an original way through which one can identify the modes of leaky systems.

To conclude, it is noteworthy to report that random lasing with resonant feedback, demonstrated here for one particular system, has been observed in a large range of different systems. We have considered a variety of scatterers configurations, varied the sample dimensions and explored a large range of index contrasts. We observed lasing action for Δn as low as 0.05, in a regime of very weak scattering, with a mean free path larger than the size of the system. Lasing is phase-coherent and is therefore different from the amplification of noise described by Mujumdar *et al.* [7]. Besides, the lasing modes are reproducible and independent of the initial noise used to initiate the laser oscillation buildup. Next, these lasing modes have a small quality factor and spread over the whole system. This precludes a scenario based on prelocalized modes [8]. Our results suggest a completely different mechanism responsible for the coherent feedback. A single leaky mode of the passive system is selected by the gain and serves as the first lasing mode. This result is in favor of recent theoretical predictions [18], stating that even in the regime of strong spectral overlap, the lasing modes are the individual quasimodes of passive systems described by a non-Hermitian Hamiltonian. We also checked systematically that the modes are not modified significantly by the nonlinear effects due to the gain [19]. An interesting output of these results is to exhibit quasimodes via the knowledge of the spatial distribution of the lasing mode, in a system where spectral selection is impossible because of strong modal overlap. This opens up the possibility of studying the biorthogonal eigenmodes in the transition from closed to fully open scattering systems [20,21]. We have focused on the single-mode lasing regime. Multimode lasing in such leaky systems will be the subject of future investigation. Finally, this work has been restricted to homogeneous distribution of the gain over the whole system. Local or heterogeneous pumping of a weak scattering system is currently under investigation [22].

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