Amplified Extended Modes in Random Lasers

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We report on a new random laser phenomenon that gives rise to narrow emission modes without requiring optical cavities. Sharp emission peaks are observed experimentally over a broad range of scattering strengths and analyzed in numerical calculations. We find that the introduction of exponential gain in a multiple light scattering process strongly increases the importance of very long light paths. Such long paths are rare and often neglected in passive disordered materials but we show that they can dominate the emission spectrum from an amplifying disordered system.

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Light waves in amplifying disordered materials are subject to a diffusive transport with gain. It was predicted theoretically by Letokhov that one can obtain laserlike emission from such materials using nonresonant feedback via multiple scattering of light [1]. This phenomenon has been first observed in strongly scattering powders [2]. The emission from such systems was shown to become narrow banded above a threshold and to exhibit laser spiking [3]. This phenomenon, also known as diffusive random lasing [4], has now been observed in a variety of materials such as powdered laser crystals [2,3], microparticles in laser dye solution [5], ceramic materials [6], and liquid crystal-dye infiltrated porous glass [7]. The phenomenon has been treated analytically [8] and through Monte Carlo simulations [9] to understand the temporal and spectral properties. The photon statistics of the emission from random lasers were calculated to be Poissonian above threshold even in the regime of diffusive scattering, which means the emission can be coherent without interference feedback [10]. Interesting resonant lasing effects have been observed in related complex systems like dye-doped cholesteric liquid crystals [11] and amplifying organic films [12,13].

Interference effects can survive random multiple scattering leading to phenomena like, for instance, coherent backscattering and strong (Anderson) localization of light [14]. Coherent backscattering has been observed to survive the introduction of optical gain in random systems [15]. Lasing is often associated with interference (coherent feedback) and recent experiments on zinc oxide powder aimed at combining random lasing and strong localization [16,17]. The combination of strong localization and gain has also been discussed theoretically, mainly for one dimensional [18] and two dimensional [19] systems. Strong localization in 3D random systems requires very strong scattering and is much more difficult to achieve than localization in lower dimensions [20].

In this Letter we report on the observation of a phenomenon in amplifying random systems that leads to narrow emission spikes and that does not require interference effects or localized modes. We will show both experimentally and numerically that the introduction of exponential gain in a finite-size random system can amplify the importance of rare long light paths to such extent that they dominate the emission spectrum. The phenomenon is a direct consequence of the amplification of noise in the path length distribution in random systems and is a general property of any random walk model with coherent gain. We therefore expect it to play an important role in most amplifying disordered materials studied in the context of random lasing.

The scattering strength of a disordered optical material is determined by its transport mean free path ℓ, defined as the length scale over which a propagating wave loses its memory of propagation direction. A material is in the multiple scattering regime when ℓ is considerably smaller then the system size L. In that case the transport of light can be described by a random walk with step length ℓ. We investigated the emission from a random laser system of which the scattering strength could be varied over a broad range in the regime of multiple scattering. We performed experiments on particle suspensions in a rhodamine 6G-methanol solution. (Dye concentration is 5×10^{-3} mol/l.) Measured amounts of zinc oxide powder were suspended into the solution. The suspension was shaken in an ultrasonic bath for 30 min to prevent coagulation of the particles. We determined the transport mean free path of our samples from measurements of the diffusion constant. For further details of this technique, see Ref. [7]. The transport mean free path of the samples was calculated using $D = v\ell/3$, with v the transport velocity [21]. We prepared 20 sets of samples in the range from $k\ell = 35$ to $k\ell = 5800$, with k the wave vector.

The samples were excited by the frequency-doubled output ($\lambda = 532.8$ nm) of a mode-locked Nd:YAG laser (2.5 Hz repetition rate, 25 ps pulse width). The pump beam was focused on the sample surface to a diameter of 67 μm. Single shot emission spectra were recorded during each excitation pulse, using a peltier-cooled CCD array coupled to a monochromator. The spectral resolution of the setup was measured using a calibrated mercury lamp and was found to be 0.25 nm. On every sample 100 individual spectra were taken and stored independently for analysis.

Figure 1 shows experimentally observed single-shot emission spectra for two cases (a) $\ell = 87.8$ μm, and (b) $\ell = 538$ μm. The excitation energy was, respectively, 8 and 21 μJ. The spectra show a distinct structure made up of several sharp peaks on top of a broader background. The location of the sharp peaks changes from pulse to pulse. The bandwidth of each peak is about 0.25 nm; equal to the spectral resolution of the setup. The height of the sharp peaks is far above the noise level in the measurement. As a control measurement we used pure dye solutions of the same concentration and observed no spectral spikes in any emission direction.

Narrow emission peaks from random systems with gain have been previously reported for ZnO powders and suspensions in dye solution in the range $k\ell \sim 5$–14, and were interpreted in terms of Anderson localization effects [16,17]. In 3D random systems, Anderson localization is a phase transition phenomenon in the disorder parameter $k\ell$ [20]. The occurrence of localized modes depends therefore in a critical way on $k\ell$ and, in particular, localized modes are expected when $k\ell \leq 1$. We found that the sharp emission peaks appeared over the complete $k\ell$ range that we investigated, $k\ell = 35$–5800, that is over more than 2 orders of magnitude of the disorder parameter. At the higher values of $k\ell$ we observed a minor increase of the pump threshold. Thus, the $k\ell$ dependence of the narrow modes was weak and inconsistent with a phase transition effect like Anderson localization.

Puzzled by these experimental results we decided to look further into this phenomenon. We modeled light transport through these random amplifying systems by a random walk using a Monte Carlo simulation technique. Detailed analysis of the random walk process with amplification then revealed the origin of the spectrally narrow emission modes. The calculation was performed in two stages. The excitation was carried out by performing a Monte Carlo simulation of the three dimensional random walk of photons at 532 nm wavelength through the sample, accompanied by their absorption by the dye. The absorption cross section of the dye at 532 nm was taken $\sigma_{abs} = 3.7 \times 10^{-16}$ cm^2 [22] and the dye concentration, transport mean free path, sample geometry, and thickness were taken equal to the values used in the experiments. The excitation light was injected in the front sample interface. The total number of photons launched into the sample was determined by the single shot excitation energy. The excitation leads to a local population inversion, that was recorded as a function of depth and transverse direction inside the sample. Azimuthal symmetry around the sample normal was used as in the experiments. In this way a realistic picture of the excited sample was obtained, including the spatial distribution of the excitation and hence of the gain.

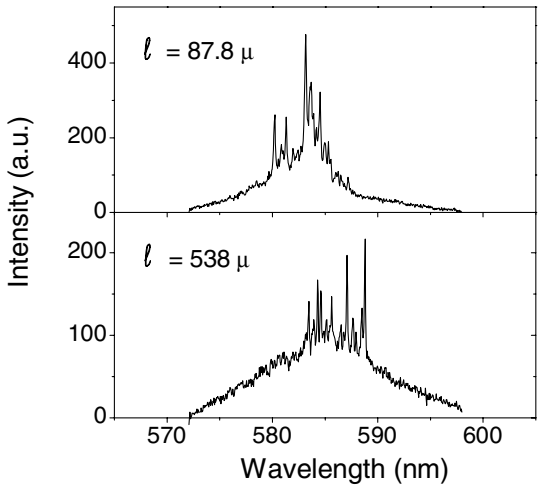

FIG. 1. Experimentally observed emission spectrum from suspensions of ZnO in Rh6G. Narrow emission modes are seen from modest and even relatively weakly scattering samples, with mean free path $\ell = 87.8$ μm (upper panel) and $\ell = 538$ μm (lower panel). The sample dimensions are $1 \times 1 \times 1$ cm.

The process of fluorescence and subsequent diffusion and amplification was simulated in the second stage. The position and the associated wavelength λ of each spontaneous emission event were picked up from the cumulative probability distributions constructed by using the spatial gain profile and the normalized fluorescence spectrum of the dye. The photon was then allowed to perform a three dimensional random walk through the active random medium that included both stimulated emission and (minor) reabsorption. The wavelength dependence of the emission and absorption cross sections $\sigma_{em}(\lambda)$ and $\sigma_{abs}(\lambda)$ was taken from literature [22]. The local concentration $N_0(\vec{r})$ and $N_1(\vec{r})$ of the dye molecules in respectively the ground and excited state were recorded and updated continuously during the simulation whenever a photon was emitted or absorbed by a dye molecule. In this way the local population dynamics were taken into account and the competition between different paths was hence included. After complete deexcitation of the sample, spectra were constructed from the recorded values of the number of emitted photons and their wavelength, maintaining a spectral resolution of 0.25 nm.

In Fig. 2, we plot calculated single shot spectra for two values of the mean free path: (a) $\ell = 87.8$ μm and (b) $\ell = 538$ μm. The excitation energy and sample parameters were the same as for the experiments reported in Fig. 1. Narrow emission modes are clearly observed as in the experiments. Moreover, the $k\ell$ dependence in the simulation in the range $k\ell = 35$–5800 reflects the $k\ell$ dependence observed in the experiments.

A Monte Carlo simulation provides access to the history of each spontaneously emitted photon and subsequent random walk which allowed us to find the origin of the sharp peaks in the emission spectrum. The broad background observed in Fig. 2 originates from a large number of spontaneous emission events that performed a random walk and were moderately amplified. On the contrary, each individual sharp peak in Fig. 2 corresponds to a *single* spontaneous emission event that followed an

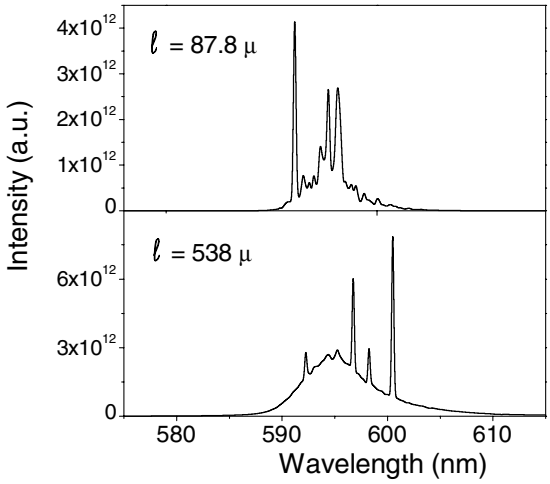

FIG. 2. Calculated emission spectra for the same sample parameters and excitation energies as in the experiments reported in Fig. 1. Sharp emission modes are seen from samples with $\ell = 87.8\ \mu$m (upper panel) and $\ell = 538\ \mu$m (lower panel). Each sharp peak can be identified in the Monte Carlo simulation as originating from a rare long light path that was amplified exponentially.

extremely long light path through the sample and was consequently strongly amplified. Such long light paths are very rare and therefore have a negligible contribution to the net transport in passive systems. The introduction of exponential gain causes a dramatic change in the importance of such long paths.

To obtain more insight into the origin of this phenomenon it is instructive to analyze the path length distribution of the random walk inside the samples. Figure 3(a) shows the probability distribution of the path lengths of the spontaneously emitted photons in the absence of gain, as calculated in our Monte Carlo simulation, for one single laser shot. The overall decay of the distribution is exponential and determined by the system size and mean free path. Noise is evident in the tail of the distribution. This noise is not caused by numerical artifacts but is due to the finite number of excited atoms after a single laser shot. This will always cause noise somewhere in the tail of the path length distribution. The noisy range in the distribution corresponds to low-probability long light paths which are irrelevant in the passive case due to the exponential falloff of the distribution function.

The situation changes when gain is introduced. Figures 3(b)–3(d) show the total intensity that paths of length N contribute to the emission, plotted versus path length. A longer path will accumulate more gain and hence contribute more to the total emission. We can clearly see in Figs. 3(c) and 3(d) that at a certain value of the excitation energy the noise in the tail of the path length distribution is amplified to such an extent that it starts to provide a dominant contribution to the total emission intensity. The natural cutoff to $I(N)$ at $N \rightarrow \infty$ is given by gain saturation. This is automatically taken

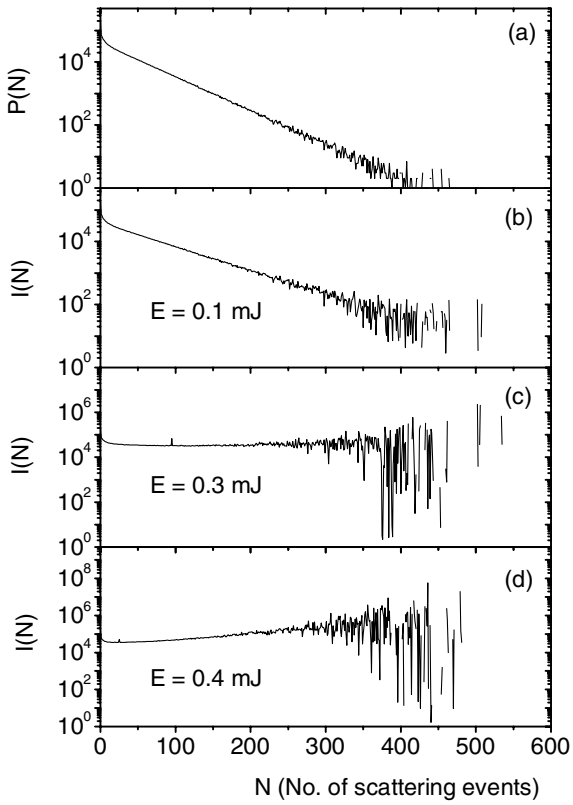

FIG. 3. (a) Calculated path length distribution of the spontaneously emitted photons. Sample dimensions: 10 mm × 10 mm × 100 μm (slab geometry), $\ell = 10\ \mu$m. (b)–(d) Intensity contribution to the emission spectrum as a function of number of scattering events for different values of the excitation energy E: (b) 0.1 mJ, (c) 0.3 mJ, and (d) 0.4 mJ. The noise in the path length distribution is amplified to such an extent that it contributes in a dominating way to the total emission.

into account in our Monte Carlo simulation as each atom is deexcited after it contributes to a stimulated emission event. The maximum amplification is therefore limited by the total number of excited dye molecules. In the case of Fig. 3(c) the gain compensates almost exactly the exponentially decaying path length distribution, yielding a nearly horizontal line that ends in a strong noisy pattern. This is exactly the situation in which the system is at threshold, defined as the excitation at which the average gain at a specific wavelength equals the overall losses (diffusion out of the sample).

The noise in the path length distribution is directly responsible for the narrow peaks in the emission spectrum. We directly observe in the Monte Carlo simulation that the highest noise spikes in Figs. 3(c) and 3(d) correspond to single spontaneous emission events that, by chance, followed a very long light path in the sample and hence picked up a very large gain. Each of the high spikes can be associated to a specific peak in the emission spectrum. The wavelength at which these modes appear is determined by the wavelength of the spontaneous emission event that originated the mode. The subsequent

stimulated emission preserves a narrow emission spectrum leading to a narrow emission peak.

The phenomenon that we have described in this Letter is generally valid for a random walk process with coherent gain starting from spontaneous emission. It should therefore apply to a broad range of amplifying disordered systems, provided the system is of finite size and in the multiple scattering regime, close to or above threshold. Whether the sharp emission peaks are observed in an actual experiment will depend on the experimental conditions. In this phenomenon, shot to shot spectra are intrinsically different due to the inherent randomness in the spontaneous emission process. Single shot experiments are therefore essential to observe the effect. This makes it distinct from interference effects that rely on coherent feedback from the system boundaries or from localized modes.

The phenomenon depends not critically but only weakly on the disorder parameter $k\ell$. The $k\ell$ dependence is that of a diffusive random laser and is additionally weakened since the excited volume increases upon increasing ℓ. (The excitation light is subject to the same diffusion process as the emission.) It would be interesting to examine if this phenomenon plays a role also in solid samples like zinc oxide, from which narrow emission peaks have been observed and shot to shot spectra were reported to be indeed distinct [23]. The emission spectra observed from amplifying (organic) films exhibit no shot to shot changes [12,13,24] and are therefore not due to the phenomenon described in this Letter. These materials have a very different geometry (thin films with large mean free path) so that the lasing mechanism is expected to be different.

The random laser modes that we have found here rely on a class of light paths that is often neglected in random systems. Whereas in the case of Anderson localization light is confined in a small region of space of dimensions determined by the localization length, the phenomenon that we have described here depends on the longest light paths that are populated in the system. These are therefore light paths that probe a large volume of the sample and hence are extended modes. These modes have a very long residence time (and so a huge amplification) whereas the chance to excite them is very small. We believe that our findings open up several possibilities for future research in the field of random lasers. Interesting topics could involve the quantum coherence properties of the observed emission modes and their statistical behavior.

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