Frequency behavior of coherent random lasing in diffusive resonant media

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Abstract

We investigate diffusive propagation of light and consequent random lasing in an amplifying medium comprising resonant spherical scatterers. A Monte-Carlo calculation based on photon propagation via three-dimensional random walks is employed to obtain the dwell-times of light in the system. We compare the inter-scatterer and intra-scatterer dwell-times for representative resonant and non-resonant wavelengths. Our results show that more efficient random lasing, with intense coherent modes, is obtained for a system with intra-scatterer gain. This is also coupled with a larger reduction in frequency fluctuations. We find that such a system can yield almost thresholdless random lasing. Inspired by these results, we discuss a possible practical situation, based on a monodisperse aerosol, wherein frequency controlled coherent random lasing can be obtained. Since our analysis essentially investigates transport of intensity, the results are relevant to coherent random lasers under nonresonant feedback.

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1. Introduction

Random lasers are intriguing optical systems that rely on the confluence of optical amplification and concurrent multiple scattering to produce radiation with unique characteristics [1,2]. Essentially, by using scattering of light to increase its interaction time with an inverted medium, stimulated emission can be enhanced [3]. The phenomenon is termed a ‘laser’ because of the threshold behavior it exhibits as the excitation energy increases, akin to a conventional resonator-based laser. This threshold is manifested when the loss in the system is overcome by the gain, which is boosted by the nonresonant feedback of the light within the system [4]. At that point, the emission intensity is seen to diverge [5]. Concurrently, the bandwidth is also observed to collapse, leading to a stable, single-mode narrowband emission [6]. Nonresonant feedback relies on the transport of intensity in the system, and can be studied under the diffusion approximation [4,7], or using the radiative transfer equation [8]. Intriguingly, coherent random lasing, comprising sub-nanometer bandwidth modes, is also reported from disordered amplifying systems [1,2]. Under strong scattering conditions, localized modes in the system yield such coherent modes [9]. In the presence of weaker scattering, such modes can originate from either of the feedback mechanisms, namely, resonant feedback or nonresonant feedback. Resonant feedback relates to the Fabry–Perot resonance that is excited between two nanoparticles, yielding a set of equispaced coherent modes [10]. On the other hand, ultranarrowband modes from nonresonant...
feedback originate from modes extended over a substantial portion of the gain volume, that are excited by spontaneous emission that comprises the initial fluorescence. Due to the adequate gain, these modes realize a large amplification and are emitted as intense modes [11]. In such systems, the underlying inversion dynamics manifest as strong fluctuations in the coherent and incoherent intensity [12]. Under strong excitation, a large number of these modes are excited to provide spectral averaging, leading to reduced coherence [13]. Theoretically, spectral features of nonresonant systems can be explained using intensity transport [14], while the resonant feedback is understood with the help of field feedback using finite difference techniques [15,16]. Over the years, many studies have reported the occurrence of random lasing and its implications in diverse systems [17–21].

A particular parameter of interest for any optical source is its emission frequency. The emission frequency of coherent random lasers exhibits strong fluctuations. In the scenario of nonresonant feedback, the frequency is determined by the spontaneous emission events which excite the extended modes. The high gain present in the system skews the contribution of these extended modes, leading to coherent emission. Hence, the spectra show chaotic fluctuations from pulse to pulse [22], essentially reproducing the fluctuations in spontaneous emission. This fact is responsible for the lack of attempts to exploit the random laser in real applications. In order to enhance applicability of the random laser, these frequency fluctuations need to be controlled. Clearly, this control cannot be brought over the exciting spontaneous emission event, but one could possibly attune the lifetime of the corresponding extended mode excited by this event. Modal lifetimes are determined by the scattering cross-section $\sigma_{\text{sc}}$, which is, by definition, a function of the wavelength. However, in conventional scatterers like dielectric nanoparticles, the wavelength sensitivity across the narrow emission band of the gain medium is rather weak. As a viable alternative, the usage of resonant scatterers can make these cross-sections wavelength-sensitive over the narrow band of interest. Indeed, the participation of resonant scatterers in mesoscopic phenomena has been well-studied in recent times. The phenomena of light diffusion and light localization have been shown to be influenced by the presence of resonant scatterers [23,24]. The wavelength-sensitivity of Mie resonances of microspherical scatterers has been exploited to achieve tuning of incoherent random lasers in systems known as photonic glasses [25]. Recently, we have numerically examined the phenomenon of coherent random lasing in a diffusive system comprising monodisperse, resonant scatterers [26]. The gain was assumed to be between the scatterers, like a conventional random laser. We have shown that such systems exhibit persistent coherent random lasing in the bands dictated by the resonances, and that the frequency fluctuations of the system are limited to this resonance band. In this paper, we investigate the dwell-time of light in the random laser in two scenarios of intra-scatterer gain and inter-scatterer gain and its consequences on the spectra.

This paper is organized as follows. After a brief description of the diffusive resonant system, we discuss the inter-scatterer and intra-scatterer dwell-time distributions therein, and the frequency behavior of the spectra from the two systems with inter-scatterer and intra-scatterer gain. Next, we discuss the case of dense scatterers (volume fraction 10%) where the dwell-times are made comparable, and the threshold excitation is compared in the two systems. Finally, a physically realistic sample is proposed wherein an aerosol-based coherent random laser is analyzed for possible frequency control.

2. Diffusive resonant media

The phrase ‘diffusive resonant media’ refers to an ensemble of multiple scatterers, each one of which has a size parameter large enough to sustain resonances. The sample is such that light propagation can be modeled as diffusive, implying the scatterers are well-separated from each other and do not interact coherently. Although this premise is motivated totally from computational convenience, experimentally it has been observed that even an ensemble of touching microspheres exhibits diffusive propagation [25]. Furthermore, the study does not encompass systems in which collective resonances over several scatterers are excited.

The Mie resonant scattering cross-section profile of a single scatterer results in a two-fold effect on light interaction. A resonant scatterer traps light inside for a time determined by the quality factor of the resonance, and hence the lifetime of the corresponding mode is already enhanced by this interaction. Furthermore, the enhanced scattering cross-section $\sigma_{\text{sc}}(\lambda)$ at the same wavelength leads to a reduced mean free path, thus leading to a larger number of scatterings. In the presence of multiple scattering, both these attributes combine to produce an enhancement in the lifetime of the light wave. A system with gain outside the scatterers will not be able to exploit the enhancement due to the intra-scatterer residence. On the contrary, a system with intra-scatterer gain benefits from both the attributes, and can be
expected to exhibit interesting features in the emission. To study such a system numerically, an accurate estimate of the dwell-times, involving both the intra-scatterer dwell-time ($\tau_{\text{intra}}$) and the inter-scatterer dwell-time ($\tau_{\text{inter}}$) that arises from multiple scattering, is necessary. To that end, a Monte Carlo algorithm for photon propagation relying on known scattering parameters like the mean free path $\ell^*$, the sample size $L$, the resonance quality factor $Q$ can be employed. The photon propagation algorithm has already been successfully utilized in modeling intensity transport of random lasers [11,14,27].

We describe here only the essence of the Monte Carlo algorithm, and complete details of the same are in Refs. [14] and [26]. In short, the Monte Carlo algorithm essentially calculates the time spent by the light wave in the multiple scattering system, whose scattering parameters are exactly known. The light wave is modeled as a packet of photons that propagate through a disordered environment, via three-dimensional random walks. During the excitation phase of the computation, these photons are assumed to be pump photons and they are absorbed in proportion to their dwell-time in the system, which is consequently excited. Subsequently, the system relaxes via the generation of spontaneous emission photons, which comprise the fluorescence. These photons also propagate through random walks, and experience gain in proportion to their dwell-time. By tracking this dwell-time, the consequent gain and the concurrent inversion dynamics are calculated. Finally, spectra are constructed by appropriately binning the amplified photons according to the wavelength. The contribution of the resonances is invoked only during the relaxation phase, since the wavelength dependence of only the emitted light is under investigation.

Fig. 1 shows two experimentally measured spectra and two calculated spectra from a conventional coherent random laser without any resonances. Characteristic frequency fluctuations that originate from the spontaneous emission photons are well reproduced in the calculations. Note that the spectral band in the experiments is obviously narrower ($\sim$12 nm) since it is limited by the apparatus. Clearly, coherent modes exist at any frequency in the range where the fluorescence cross-section is large, with strong fluctuations in this range.

In the computational domain, the resonant effects are embedded in the transport mean free path $\ell^* = \frac{1}{\Phi} (\frac{d}{Q_{\text{sca}}})$, where $\Phi$ is the volume fraction of scatterers, $Q_{\text{sca}}$ is the Mie scattering efficiency, $d$ is the scatterer diameter and $g$ indicates the scattering anisotropy. For calculating the time spent in the scatterers, the Mie resonances observed in the $Q_{\text{sca}}$ profile were fit by a Lorentzian, and the quality factors thereof were measured. From the Lorentzian profile and the quality factors, the decay times of photons for various wavelengths in the resonance band were computed.

Fig. 2 depicts the various parameters of the resonant system. These numbers were calculated for a system comprising spheres of diameter 1.09 $\mu$m, with a refractive index contrast (with ambient medium) of $n = 1.8$. The top plot depicts the Mie resonance peaks in the scattering efficiency profile (blue) within the band of interest (the Rhodamine 6G emission profile, shown in red). The height of these peaks determines the mean free path, and the width on the frequency axis determines the quality factor. Accordingly, the resonant peaks, extracted by fitting Lorentzians to the profile, are shown in black. The bottom plot shows the mean free path $\ell^*$ having dips at the positions of the Mie resonances, indicating stronger scattering of light. This enhances the number of scattering events encountered by the fluorescent photons, as seen in the black profile. At each event, the photon spends some residence time inside the scatterer. ($\tau_{\text{inter}}$) is obtained from the total pathlengths of the photons, while ($\tau_{\text{intra}}$) is calculated by the cumulative time delays within the multiple scatterers. The simultaneous enhancement in ($\tau_{\text{inter}}$) and ($\tau_{\text{intra}}$) will lead to suppressed diffusion at the resonant wavelengths.

![Fig. 1. (A) Experimentally observed and (B) numerically calculated spectra from a nonresonant random laser, showing pulse-to-pulse frequency fluctuations. Neither system has resonant properties.](image-url)
3. Results and analysis

3.1. Sparse system

With a volume fraction of 1%, the scatterers are sparsely populated in the system, resembling a conventional random laser wherein dielectric scatterers are suspended in an amplifying medium. For such a configuration, Fig. 3 shows the calculated dwell-times, wherein the top plot depicts the $\tau_{\text{inter}}$, and the bottom plot shows the $\tau_{\text{intra}}$. Two wavelengths are chosen for analysis, the resonant $\lambda_{\text{res}} = 559.8$ nm, and the nonresonant $\lambda_{\text{nres}} = 563.3$ nm. The $\tau_{\text{inter}}$ shows a slight increase for $\lambda_{\text{res}}$ due to the stronger scattering cross-section. This difference is only modest, because the large number of scattering events is compensated by the small $\ell'$, thus reducing the disparity in $\tau_{\text{inter}}$. The bottom plot for $\tau_{\text{intra}}$ discusses an additional wavelength $\lambda = 560.3$ nm, which lies near the half-maximum of the resonance. (Note the difference in the X-axis scale). Here, evidently, the disparity between the $\lambda_{\text{res}}$ and $\lambda_{\text{nres}}$ is large, clearly because the nonresonant light is not sustained in the scatterer.

It is instructive to observe the effect of this temporal distribution on the spectra. We studied two scenarios, one in which the gain was assumed to be in between the scatterers, and the other wherein the gain was inside the scatterers only. Two representative spectra, out of the calculated 75 spectra, are shown in Fig. 4. Persistent lasing was seen in both the cases, but striking differences were also observed. In the case of inter-scatterer gain (black profile), in addition to the coherent
scattering, the dwell-time distribution naturally is altered. Here, we discuss such a system with a volume fraction of 10%, wherein the mean free paths are of the order of 5–7 μm. For stronger volume fractions, the situation leads to correlated (touching) scatterers, and the scattering parameters need to be calculated differently. Nonetheless, we believe the qualitative inferences of this study can be carried over to larger scattering strength as well. Fig. 5(a) and (b) shows the $\tau_{\text{inter}}$ and $\tau_{\text{intra}}$ in the system. Here, due to the largely reduced mean free path, the $\tau_{\text{intra}}$ decreases compared to the case with sparse scatterers. In contradiction, the $\tau_{\text{intra}}$ shoots up due to the increased number of scatterings, and the two dwell-times are comparable.

Fig. 5(c) further depicts the behavior of the peak intensity in the spectrum as a function of the excitation energy.

### 3.2. Dense scatterers

The earlier system with the said parameters was essentially a sparse scattering system (volume fraction 1%), leading to a larger $\tau_{\text{inter}}$ and hence is attuned for inter-scatterer gain. A practical system with intra-scatterer gain can allow denser packing of the amplifying scatterers, minimizing the mean free path, to realize a compact random laser. In the case of dense scatterers, the dwell-time distribution naturally is altered. Here, we discuss such a system with a volume fraction of 10%, wherein the mean free paths are of the order of 5–7 μm. For stronger volume fractions, the situation leads to correlated (touching) scatterers, and the scattering parameters need to be calculated differently. Nonetheless, we believe the qualitative inferences of this study can be carried over to larger scattering strength as well. Fig. 5(a) and (b) shows the $\tau_{\text{inter}}$ and $\tau_{\text{intra}}$ in the system. Here, due to the largely reduced mean free path, the $\tau_{\text{intra}}$ decreases compared to the case with sparse scatterers. In contradiction, the $\tau_{\text{intra}}$ shoots up due to the increased number of scatterings, and the two dwell-times are comparable.

Fig. 5(c) further depicts the behavior of the peak intensity in the spectrum as a function of the excitation energy.
intensity. In the case of inter-scatterer gain (black curve), a threshold is observed at ~0.3 μJ, after which the output intensity grows rapidly. The bottom inset shows a representative spectrum at 0.4 μJ, indicating the coherent modes on the broadband pedestal. With higher excitation, the modes grow at the cost of the pedestal, creating a larger number of intense coherent modes. With intra-scatterer gain (red curve), the intensity rises rapidly even with the smallest excitation. This is due to the excessive residence times of the resonant photons in the scatterers which emphasizes the gain of the modes. Thus, the absorbed energy is rapidly channeled into the coherent mode by the system. This also suppresses the incoherent pedestal that is created by the fluorescence. Consequently, very clean, ultra-narrowband coherent random lasing modes are seen. A representative spectrum at excitation energy of 0.2 μJ is shown in the top inset. Upon further excitation of the sample, gain saturation of the coherent peaks occurs, and the maximum intensity tends to saturate. This analysis indicates that very low threshold, or almost thresholdless lasing, can be obtained from a dense ensemble of amplifying resonant scatterers.

4. A practical proposal: an aerosol random laser

We propose a potentially realistic system which can be used to observe frequency control in coherent random lasing using Mie resonances. Towards the requirement of high gain, the right material remains a lasing dye dissolved in an alcohol, which has gain cross-sections $\sigma_g \sim 10^{-16}$ cm$^2$. A possible mechanism to create amplifying scatterers with the above $\sigma_g$ would be to create microdroplets of the said dye solution. Such a microdroplet will have a refractive index contrast of 1.329 (methanol microdroplet in air), and will offer a high gain cross-section for the light resident inside it. An ensemble of such microdroplets suspended in air, technically an aerosol, will act as a disordered medium with high intra-scatterer gain. We have recently experimentally demonstrated such an aerosol-based coherent random laser, created out of dye microdroplets with a larger diameter [28]. Frequency control was not seen in that system because of the strong polydispersity in the aerosol. Here, we model the emission of such a system to observe the frequency behavior. Fig. 6(A) shows the $Q_m$ (blue curve) from a spherical scatterer with diameter 3.6 μm, and $n = 1.329$. Compared to the earlier spectra, the Mie resonances in these scatterers are broader due to the lower refractive index. Two typical spectra from this system pumped at 7 μJ are shown in red. The effect of broadband nature of the resonance is seen clearly. Multimode emission is observed at each resonance location, instead of a single mode seen in earlier particles. These modes fluctuate within the broad resonance band, and tend to average out with increasing excitation intensity. A polydisperse system was also simulated by assuming 12% spread in the diameter of the microscatterers. The black spectrum depicts the behavior of the polydisperse system at 30 μJ, wherein the signatures of the resonance are washed out. The larger excitation energy required to generate coherent peaks is also a consequence of the polydispersity. Here, the frequency behavior is similar to a conventional random laser. A summary of the frequency fluctuations can be obtained by the histogram of the lasing frequencies over 75 spectra. Fig. 6(B) shows the computed histogram from the polydisperse aerosol (gray columns), and the monodisperse aerosol (red columns). The polydisperse system shows a
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