Coherent random lasing in diffusive resonant media

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Abstract. We investigate diffusive propagation of light and consequent random lasing in a 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 when the gain is present inside the scatterers. Further, a larger reduction in frequency fluctuations is achieved by the system with intra-scatterer gain.

Keywords: Random lasers, Wave propagation in random media, Scattering, Light diffusion

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INTRODUCTION

Random lasers rely on amplification and multiple scattering to create coherent emission from disordered systems[1, 2]. Under certain conditions of gain and disorder, coherent modes are generated without the need of a resonator[3]. The underlying complex inversion dynamics, dictated by overlapping modes and consequent mode competition, leads to fluctuations in the coherent and incoherent intensity[4]. Furthermore, the lasing wavelength of the random laser is determined by the spontaneous emission events that trigger amplified extended modes within the system, resulting in strong fluctuations in the emission from pulse to pulse[5]. To bring the random laser closer to real applications, some control needs to be achieved on the emission wavelength. While a spontaneous emission event is hard to control, one could possibly alter the lifetime of the corresponding extended mode. The lifetimes of the modes are determined by the cross-sections, which can be altered using resonant scatterers. The possible contribution of resonant potentials in localization phenomena has been pondered upon from very early days[6]. Indeed, in more recent experiments, light diffusion and localization has been shown to be influenced by the presence of resonant scatterers[7, 8]. Even in active systems, the longer lifetimes of modes have been exploited in resonance-driven incoherent random lasers to tune the peak wavelengths by exploiting the Mie resonances of spherical scatterers[9]. It is, therefore, of interest to study the contribution of resonant scatterers in the regime of coherent random lasing.

The scattering cross-section profile of a single spherical scatterer exhibits a rich structure consisting of a multitude of Mie resonant peaks, which create two simultaneous effects on light interaction. Firstly, the enhanced scattering cross-section implies that a light wave is scattered more often, while the quality factor of the resonance indicates the excessive time the light is trapped in the resonator during every scattering. In a multiply scattering environment, both these features contribute to the lifetime of the light wave. If optical amplification is to be introduced in this environment, a question arises as to whether the gain should be inside the scatterers, or in the ambient medium. An accurate estimate of the dwell-times requires a calculation based on the scattering parameters like the mean free path, the sample size, the resonance quality factor etc, which can be accomplished by a Monte Carlo algorithm for photon propagation. Such an algorithm has already been successfully utilized in modeling of random lasers[3, 10], apart from other numerical techniques[11]. Using the same algorithm, we have recently addressed the effects of single particle resonances when gain is present in the ambient medium outside the scatterers[12]. In this paper, we numerically investigate the dwell-time of light in the random laser, and its consequences on the spectra without making any presumptions about the location of the gain.

MONTE CARLO SIMULATIONS

In short, the simulation consists of two parts: excitation and the subsequent emission. During the excitation, the pump photons undergo a random walk through the sample and create a local population inversion which is recorded as a function of depth. In the subsequent stage, the random walks of fluorescence photons are implemented, taking into account the gain they gather and the concurrent inversion dynamics in order to yield the output spectra. Complete details of the numerical simulation are in Reference [3] and [9]. Figure 1 shows two experimentally measured spectra from a nonresonant coherent
random laser, and two spectra obtained from the said algorithm. The characteristic spectral fluctuations are well reproduced in the calculations. The larger spectral width of the simulations originates from the fluorescence cross-section, while the experimental bandwidth is limited by the apparatus. Notably, the coherent modes populate the region where the fluorescence cross-section and stimulated emission cross-section are larger. Within this region, the wavelength of the modes shows strong fluctuations.

In the current computations, the resonant effects are invoked via the transport mean free path $\ell^* = \frac{1}{\Gamma g \left( \frac{2d}{3Q_{sca}} \right)}$, where $\Phi$ is the volume fraction of scatterers, $Q_{sca}$ is the Mie scattering efficiency, $d$ is the scatterer diameter and $g$ parametrizes the scattering anisotropy. For the time distribution calculations, the quality factors of the resonances were calculated, and the decay times of photons for various wavelengths in the resonance were computed therefrom. Figure 2 summarizes the parameters affected by scatterer resonances. These numbers were calculated for a system comprising TiO$_2$ spheres ($n = 2.4$) of diameter 1.09 μm (with a polydispersity of 1.5%), at a volume fraction $\Phi$ of 0.01 in methanol. The variation of $\ell^*$ with wavelength exhibits sharp dips corresponding to Mie resonances. Consequently, light at the resonant wavelength experiences a stronger scattering environment in the random laser. As a result, the number of scattering events encountered by fluorescent light is enhanced, as shown by the black profile. Overall, the profile follows the fluorescence lineshape of the lasing dye, punctuated by sharp peaks at the resonant wavelengths. The scattering events almost double at $\lambda_{res} = 559.8$ nm. At each event, the photon spends some residence time inside the scatterer. The total pathlengths of the photons yielded the inter-scatterer dwell-times, while the cumulative time delays within various scatterers provided the intra-scatterer dwell-times. Evidently, the stronger scattering environment leads to suppressed diffusion of light due to simultaneous enhancement in inter-scatterer dwell-time and intra-scatterer lifetime.

The calculated dwell-times are shown in Figure 3. The plot [A] shows the inter-scatterer diffusion time for two wavelengths of interest, the resonant $\lambda_{res} = 559.8$ nm, and the nonresonant $\lambda_{nres} = 563.3$ nm. A slightly larger dwell-time is evident for the resonant wavelength due to the stronger scattering. Note that the difference is not excessive, because although the number of scattering events is larger for $\lambda_{res}$, the corresponding $\ell^*(\lambda_{res})$ is smaller, which offsets the disparity in the inter-scatterer dwell-time. The middle plot shows the intra-scatterer dwell-time for the three wavelengths. (Note the difference in the X axis scale). Here, evidently, the disparity between the $\lambda_{res}$ and $\lambda_{nres}$ is understandably large. The right plot depicts the total dwell-time of light in the random laser, as would be measured in an experimental situation. Clearly, the behavior is dominated by inter-scatterer diffusion, with very small enhancement from intra-scatterer dwell-time.

We further studied the effect on coherent random lasing using these data. In one scenario, the gain was assumed to be in between the scatterers, compared to another scenario, in which the gain was assumed to be inside the scatterers only. Both systems manifest lasing modes at the same Mie resonance. Several interesting observations were made over 40 different calculated spectra, out of which two representative spectra are shown in Figure 4. In the case of inter-scatterer gain (black profile), the coherent modes exhibited a larger propensity to appear at the location of the resonance, however, modes with lower intensity also appear outside the resonance. The wavelength fluctuations are slightly lowered because
of the high-intensity modes in the resonant region. The overall spectrum undergoes bandwidth narrowing because of multiple ultra-narrow modes. In the case of intra-scatterer gain (red profile), high intensity coherent modes appear only at the resonance, with their intensity larger than in the earlier case. Furthermore, no or minimal linewidth narrowing is seen in the general profile of the spectrum. Importantly, because many photons contribute to lasing modes in this narrow resonance, the modes rapidly undergo self-averaging to yield a broader, but a more stable random lasing mode, the width of which is determined by the Mie resonance. Thus, although the inter-scatterer dwell-time is much larger than the intra-scatterer time, we obtained cleaner and more stable random lasing from the case with intra-scatterer gain. This behavior can be attributed to a larger disparity in the intra-scatterer dwell-times of \( \lambda_{res} \) and \( \lambda_{nres} \), which leads to better selectivity of gain for the extended modes. Our studies show the way towards frequency controlled coherent random lasers.

REFERENCES