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Higher order processes in random Raman lasing

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Abstract

Random Raman lasers offer a unique opportunity to study many exciting dynamics of light propagation in turbid media. One of the most notable features observed to exist in the recently discovered random Raman laser are the presence of higher order stimulated Raman scattering (SRS) processes. The higher order Stokes generation likely comes from photons that have the longest pathlengths, thus have the most gain. This makes these photons particularly likely to offer interesting insight into wave propagation effects such as coherent backscattering and optical Anderson localization. In this work, we use Monte Carlo simulations to investigate how these higher order processes occur and what properties they are expected to exhibit when considering only transport equation dynamics. This knowledge will allow us to look for deviations from this theory in future experiments to determine if wavelike properties play an active role in random Raman lasing.

1 Introduction

Random lasing has been a topic of considerable interest for nearly two decades [1–7]. Much of this interest is centered around the opportunity to study phenomena which are typically weak in a traditional scattering medium that has losses instead of gain. The effects being investigated include coherent backscattering, also known as weak localization [8–10], and Anderson localization of light, also referred to as strong localization [11–15]. These processes are studied in random lasing media due to the presence of gain. Gain acts to preferentially amplify long path length photons, those most likely to be affected by localization processes, instead of absorption which attenuates these very photons which you wish to study.

The recent discovery of random Raman lasing has opened up a new regime in which to study light propagation in turbid media [16,17]. Random Raman lasing offers several potential advantages over traditional random lasing for the study of these effects. First, random Raman lasers can be pumped with much more intense laser pulses because the medium, typically barium sulfate (BaSO_4), has extremely small linear and multiphoton absorption. This is difficult to accomplish in traditional random lasing because the medium must be able to absorb the pump wavelength. Fundamentally, stimulated emission, the gain mechanism in traditional random lasing, requires the physical absorption of photons followed by re-emission some time later, while stimulated Raman scattering (SRS) operates

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on virtual transitions and is essentially instantaneous. This fundamental difference in the processes leads to less heat being generated in the medium and allows much more powerful laser sources to be used without damaging the lasing medium. The more intense pump laser pulses used in random Raman lasing support tremendous gain, allowing for the generation of a very bright emission [18]. Second, the gain in random Raman lasing is fundamentally different than traditional random lasing. Because SRS does not involve the actual absorption of a photon the process cannot store energy unlike stimulated emission, which stores the pump photons energy until a stimulated emission process occurs or the molecule spontaneously relaxes. This transient nature of the gain manifests itself in the gain being proportional to the local intensity of the pump field. Additionally, random Raman lasing gives us an opportunity to study nonlinear optical effects in a turbid media. A better knowledge of these dynamics will help improve upon applications ranging from chemical analysis in turbid media [19–21] to stimulated Raman induced photoacoustics [22–24].

One of the most striking properties of the random Raman lasing emission previously reported is the presence of higher order SRS processes [17,18]. These processes are the result of the Stokes shifted field undergoing an additional SRS processes, and picking up an additional Stokes shift. This is illustrated schematically in Fig. 1, and the experimental data is reproduced in Fig. 2. These higher order emissions require both a large gain and a long pathlength in the medium to develop, thus they are a natural place to investigate for the role of localization type effects in random Raman lasers.

In order to study the effects of localization, we first must understand how these higher order processes could develop in the absence of wave effects such as localization. Monte Carlo simulations treat light as a pure particle, thus they are not sensitive to wave effects that are responsible for localization. In other words, Monte Carlo simulations provide us with a view of what the emission from the random Raman lasing would look like in the complete absence of localization. This picture will allow us to compare these results to future experimental results, and enable us to conclude whether or not wave propagation effects play an important role in the dynamics of random Raman lasing.

In this work, we present Monte Carlo results which study some of the most striking features of the higher order Stokes processes which have been observed in random Raman lasing. We first study the threshold behavior of these higher order modes and then move forward to discuss the temporal behavior of these dynamics in the absence of wave propagation effects. This data will allow us to better study wave propagation effects in random Raman lasing by elucidating which effects can be explained by ordinary transport equation dynamics and which effects require wave propagation to be fully described.

2 Methods & Results

To model the behavior of the higher order processes, we employ the nonlinear Monte Carlo (NLMC) simulation developed previously [25]. To summarize the method here, elastic scattering is treated by assigning a random distance to the next elastic scattering event, given by the distribution

$$\rho(d) = \mu_s e^{-\mu_s d} \quad (1)$$

where μ_s is the scattering coefficient and d is the distance traveled by the photon. Once a scattering event occurs, the scattering angle is given by the Henyey-Greenstein distribution function in terms of the anisotropy $g = \langle \cos(\theta) \rangle$, and a uniform random number, ξ , between 0 and 1

$$\cos(\theta) = \frac{1}{2g} \left[1 + g^2 - \left(\frac{1 - g^2}{1 - g + 2g\xi} \right)^2 \right] \quad (2)$$

Absorption is treated by assigning each photon to start with a weight of unity. After each step its new weight is given in terms of the absorption coefficient, μ_a , the distance traveled in that step, d , and the index of refraction of the medium, n , by

$$w' = e^{\mu_a(d/n)} w. \quad (3)$$

Each step, photons are given a probability of undergoing spontaneous Raman scattering given by,

$$P_R = 1 - e^{\beta_R(d/n)}. \quad (4)$$

Here, β_R is a measure of the strength of the Raman transition. Due to computational restraints, this parameter must be taken to be artificially large. However, over a reasonable parameter space this has not been seen to cause large deviations in the behavior of the model outside of an overall multiplicative constant in the strength of the signal. Additionally, comparison with experimental results confirm that the parameters here are in good agreement. When a photon Raman scatters it is converted from one photon type (pump, 1st Stokes, 2nd Stokes, etc.) to the next Stokes line. To include SRS in the simulation, the photon densities of each type of photon are calculated using a binning method. Once this is done the probability of a photon undergoing SRS is given by

$$P_{\text{SRS}} = 1 - e^{\beta_{\text{SRS}} \rho_R(d/n)} \quad (5)$$

where ρ_R is the photon density of the Stokes mode that the photon will scatter into if this “coin flip” is successful, and β_{SRS} is a measure of the strength of the SRS process.

To model random Raman lasing, we chose the parameters which give the best fit to the available experimental data [17]. The sample was set to be 5 mm thick and 6 mm in the transverse directions. No photons in any of the simulations were able to penetrate through the sample, thus it can be treated as semi-infinite. Photon packets encountering the side boundaries of the sample were terminated at the surface. While this is a potential source of error, the few photons which do penetrate this far are extremely unlikely to undergo stimulated processes because the photon density is so small in this region. Thus, we can safely conclude that any error incurred by this finite transverse sample approximation will

be very small compared to the stimulated contribution of the effects which we are interested in. The index of refraction was set to $n = 1.6$ to correspond to the index of refraction of BaSO_4 . The medium was given an anisotropy factor of $g = 0.6$, a scattering coefficient of $\mu_s = 200 \text{ mm}^{-1}$, an absorption coefficient of $\mu_a = 0.1 \text{ mm}^{-1}$, a spontaneous Raman coefficient of $\beta_R = 5 \times 10^{-4} \text{ mm}^{-1}$, and a stimulated Raman coefficient of $\beta_{\text{SRS}} = 1.5 \times 10^{-5} \text{ mm}^2$. The spontaneous Raman coefficient and the stimulated Raman coefficient govern the strength of their respective process in the Monte Carlo simulations. The global time step of the simulation was set to 0.1 ps and the bin size was set to 20 μm . A single incident pump pulse was used in all the simulations, however the strength of this pulse was varied. The pump pulse was Gaussian in space and time, with a full-width at half-maximum pulse length of 50 ps and a $1/e^2$ beam diameter of 1 mm. In all cases the collected signal is all the photons of a particular type that exit the sample in a reflection geometry. For every set of parameters the Monte Carlo simulations were averaged over 10 runs, each using a different initial seed for the random number generator.

The threshold behavior of the different Stokes lines is shown in Fig 3. For these runs, all the parameters described above were held constant and the number of photon packets making up the pump pulse were varied from 20,000 to 500,000 in increments of 20,000. This is akin to varying the pulse energy of the pump pulse. It should be noted that while the 4th Stokes signal is small, it is nonzero and beginning to undergo stimulated processes for the largest pump power used. One of the more surprising features of this is just how efficiently the 1st Stokes photons can be converted to these higher order modes. At the largest pump power used, 24% of the pump photons were converted to Raman photons of some kind. Of these, nearly 1 in 3 were converted into higher order Stokes modes.

The temporal dynamics of these processes are shown in Fig. 4. These are expected to be paramount to understanding the role of wave effects in random Raman lasing because localization effects will lengthen the amount of time that photons will stay in the medium. One of the interesting features that can be seen is the delay between the pump signal and the 1st Stokes, and between the 1st Stokes and 2nd Stokes and so on. It is readily seen that this delay decreases as the pump intensity is increased. The most likely reason for this is that at high pump intensities, the threshold for SRS processes occurs before the peak of the pulse. Thus, the stronger the pump pulse, the earlier that the system crosses this threshold. For sufficiently intense pump pulses, pump depletion effects can start to be seen in the scattered pump signal, and even in the 1st Stokes signal at a high enough intensity.

Furthermore, the total decay time of the pump pulse and Stokes modes does not vary significantly over the range of scattering coefficients that are reasonable for a highly scattering powder (30 mm^{-1} to 1000 mm^{-1}). This is shown in Fig. 5. In Fig. 5 it is readily visible that increases in scattering lead to increased efficiency of random Raman lasing. This is a result of the lower scattering media allowing the photons to more easily diffuse throughout, lowering the photon densities, and reducing the SRS gain.

3 Discussion

Typically higher order SRS processes are only observed in systems such as propagating high intensity pulses down a fiber or when high intensity femtosecond pulses are used in a Raman crystal [26]. The presence of these higher order modes in random Raman lasing using an unfocused 50 ps pulse is quite surprising. Furthermore, the generation of each line is surprisingly efficient, even in the absence of localization effects. In the presence of localization effects, it is logical to expect the generation of these higher order modes to be even more efficient, due to the longer interaction paths.

While an analytical approach to describing the dynamics of a 3D random Raman laser would be completely intractable, Monte Carlo simulations offer a good opportunity to understand how these dynamics fit into those associated with particle like transport equations dynamics. Such an understanding will be necessary in the search for definitive proof that optical Anderson localization can exist in a bulk 3D system. By comparing these results with future experiments, we will be able to search for deviations from transport theory that would signify the presence of wave propagation effects. Additionally, we will be able to distinguish the difference between localization effects and absorption, which has been an issue in previous searches for localization.

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Figure 1.

Conceptual drawing of higher order SRS processes where the Stokes generations undergo SRS processes themselves. Each order of the process gives the output light an additional frequency shift equal to the energy of the vibrational transition.

Figure 2.
Experimental data reported previously [17] showing the clear presence of higher order Stokes modes.

Figure 3.
Monte Carlo simulations showing the threshold like behavior of each of the Stokes lines.

Figure 4.

A Temporal distribution of the input pump pulse used in the Monte Carlo simulations. Note that the vertical scale for this is arbitrary as it depends on the strength of the input pump used. **B-F** Temporal dependence of the various Stokes lines as well as the residual pump for different strength pumps in a reflection geometry. All five plots are plotted using the same vertical scale, thus are comparable. The input pump strengths are as follows: **B** 100,000 photon packets, **C** 200,000 photon packets, **D** 300,000 photon packets, **E** 400,000 photon packets, and **F** 500,000 photon packets.

Figure 5.

Temporal dependence of the scattered pump and the various Stokes orders for various scattering coefficients. **A** $\mu_s = 30 \text{ mm}^{-1}$, **B** $\mu_s = 100 \text{ mm}^{-1}$, **C** $\mu_s = 300 \text{ mm}^{-1}$, **D** $\mu_s = 1000 \text{ mm}^{-1}$. 250,000 pump photon packets were simulated for each run and all other parameters are those used in all the other simulations.