Evidence of Anderson localization effects in random Raman lasing

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ABSTRACT

Anderson localization, also known as strong localization, is the absence of diffusion in turbid media resulting from wave interference. The effect was originally predicted for electron motion, and is widely known to exist in systems of less than 3 dimensions. However, Anderson localization of optical photons in 3 dimensional systems remains an elusive and controversial topic. Random Raman lasing offers the unique combination of large gain and virtually zero absorption. The lack of absorption makes long path length, localized modes preferred. The presence of gain offsets what little absorption is present, and preferentially amplifies localized modes due to their large Q factors compared with typical low Q modes present in complex media. Random Raman lasers exhibit several experimentally measured properties that diverge from classical, particle-like, diffusion. First, the temporal width of the emission being 1 to a few nanoseconds in duration when it is pumped with a 50 ps laser is a full order of magnitude longer than is predicted by Monte Carlo simulations. Second, the random Raman laser emission is highly multi-mode, consisting of hundreds of simultaneous lasing modes. This is in contrast to early theoretical results and back of the envelope arguments that both suggest that only a few modes should be present. We will present the evidence that suggests a divergence from classical diffusion theory. One likely explanation, that is consistent with all of these anomalies, is the presence of high-Q localized modes consistent with Anderson localization.

Keywords: Random Raman lasing, Anderson localization, Wave effects, Stimulated Raman scattering

1. INTRODUCTION

Lasers are promising to revolutionize lighting technology, with laser headlights already appearing in production automobiles. For some of these applications, the coherence of a laser is a significant hindrance. One potential class of lasers that can overcome this is random lasers.[1] Their highly multi-mode emission gives their emission properties more like a classical light bulb or a light emitting diode (LED), yet they exhibit laser-like brightness due to the presence of exponential gain.[2] In addition to promising applications, random lasers push our understanding of the physics of wave propagation. The complex intertwining between multiple scattering and nonlinear gain provide a seemingly ideal platform to study fundamental wave propagation processes, such as Anderson localization,[3] but accepted examples of Anderson localization, in the optical domain, in 3-dimensional systems remains elusive.

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Anderson localization, which was discovered by Anderson in 1958,[4] is of great interest in solid-state physics. He demonstrated that the wave-packet of an electron can be absent of diffusion in a disordered medium.[5] Elucidating the underlying physics in a medium with strong disorder provides insight on the transport of particles, including electrons and photons. Recently, Anderson localization of light in photonic lattices with disorder has drawn considerable interest.[6,7] Due to the imperfections in optical media, it is impossible to disregard disorder in any optical system. Thus, the performance of systems with strong disorder is of great importance for known optical phenomena. For example, random nano-lasing has been studied in the Anderson localized regime.[3] However, there has not been any research into the effect of Anderson localization on random Raman lasing.

Random Raman lasing,[8,9] random lasing which uses stimulated Raman scattering as the gain mechanism, is emerging as an interesting platform to study wavelike phenomena. Fundamentally, the advantage of random Raman lasing, compared to traditional random lasers based on stimulated emission, is the low absorption of the pump radiation. This allows the pump to penetrate a much larger volume than stimulated emission lasing, where strong pump absorption is required to obtain a population inversion. In addition to low absorption at the pump wavelength, random Raman lasers have virtually no absorption at the emission wavelength, allowing the lasing emission to propagate very long paths without attenuation. On the other hand, traditional lasing requires non-zero absorption for the emission wavelengths because detailed balance guarantees that stimulated emission and absorption share the same coefficient. Ultimately, this means that propagation in regions where the pump was unable to generate a population inversion suffers significant losses due to absorption. In addition to significantly reducing the losses for extended, or long path-length modes, the low absorption possible in random Raman lasing systems allows for very intense pump pulses to be used without damage to the powdered sample, leading to an extremely bright emission which is attractive as a light source.[10,11]

In this proceedings, we present several pieces evidence that point to a deviation from classical radiation transport, both of which are consistent with effects expected from the presence of extended modes and Anderson localization. First, non-linear Monte Carlo simulations have provided useful insights into the dynamics of random Raman lasing,[12,13] and have been able to reproduce all experimental features quite well with one exception. They fail to predict the very long temporal life-time of the Raman emission. Second, random Raman lasing emission has been shown to be highly multi-mode,[11] consisting of hundreds of lasing modes, while results from more rigorous theoretical calculations predicts at most a few modes are possible. This hints at multiple, essentially independent random Raman lasers are operating simultaneously, which again, points to extended, localized modes playing a role in the dynamics. It is important to fully disclose that none of these constitute direct evidence of Anderson localization, but each of them contradicts with the current theory and appear to consistent with the presence of

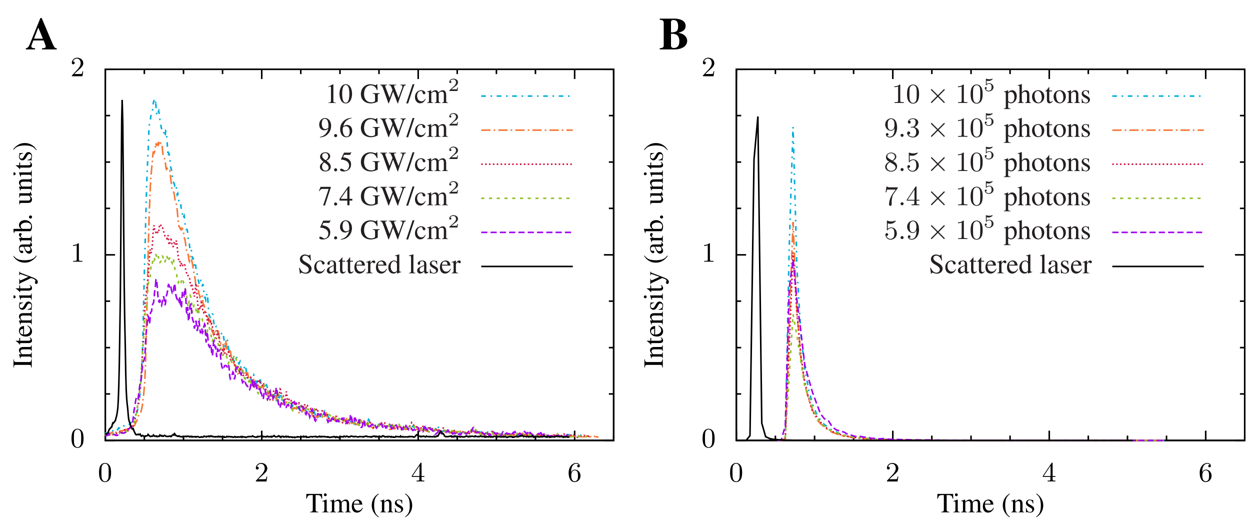

Figure 1. The temporal profile of the random Raman laser emission versus pump energy. (A) is the experimental data and (B) is the results of non-linear Monte Carlo simulations. The elastically scattered pump is shown for reference. Each trace has been normalized so that the exponential tails overlap by fitting an exponential decay and dividing each trace by the normalization coefficient. The exponential decay appears independent of pump power.

extended highly localized modes.

2. RESULTS

Barium sulfate ($BaSO_4$) was used as the active Raman medium and the source of scattering in the random Raman laser. The powder was lightly packed into a 1 cm diameter, 1 cm deep cylindrical container with the top open so that the laser never interacts with the sample container. This powder has a transport path length of 1.19 ± 0.03 μm which was measured using the coherent backscattering technique.[14] Given this very short transport path length we are likely in the strong localization regime.

A streak camera (C1587; Hamamatsu) with a fast streak card (M1952; Hamamatsu) capable of 2 ps temporal resolution was used to measure the temporal dynamics of the random Raman laser emission. The results are shown in Fig. 1. They demonstrate that significant levels of Raman light remains present in the cavity several nanoseconds after the pump pulse. This is in contrast to non-linear Monte Carlo simulations[12,13] which predict a significantly shorter emission. The parameters used in these simulations correspond to experimental data as much as possible and are an index of refraction, $n = 1.636$, a scattering coefficient, $\mu_s = 2132.5$ mm^{-1}, an anisotropy parameter, $g = 0.7428$, an absorption coefficient, $\mu_a = 0.01$ mm^{-1},[15] a spontaneous Raman coefficient, $\beta_R = 2 \times 10^{-4}$, and a stimulated Raman coefficient, $\beta_{\mathrm{SRS}} = 10^{-6}$. The anisotropy and scattering coefficients used were computed using Mie scattering theory for a 1 μm sphere of $BaSO_4$ and are in reasonable agreement with those measured. For comparison, these values produce a transport path length of 1.82 μm. The computed values were used because the simulation requires the scattering coefficient and the anisotropy

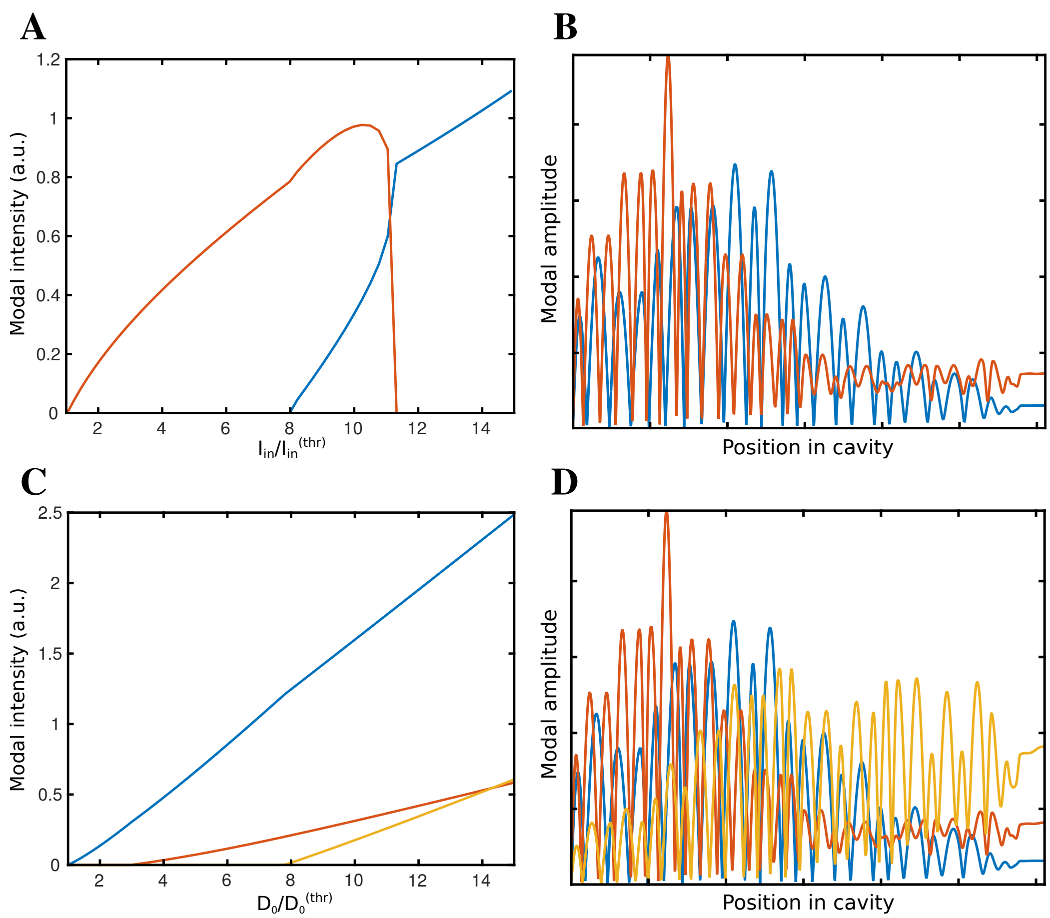

Figure 2. $L \approx 20\lambda_a$, $\lambda_a = \frac{2\pi}{40}$, the layers of the random cavity are $n = 1$, $n = 1.6$ (chosen due to n for barium sulfate), with 40 layers in total of different widths. Gain bandwidth = 4 (units of L/c). the Raman cavity is illuminated from right side. Both cavities have mirror on their left edge. Both cavities have uniformly distributed gain atoms.

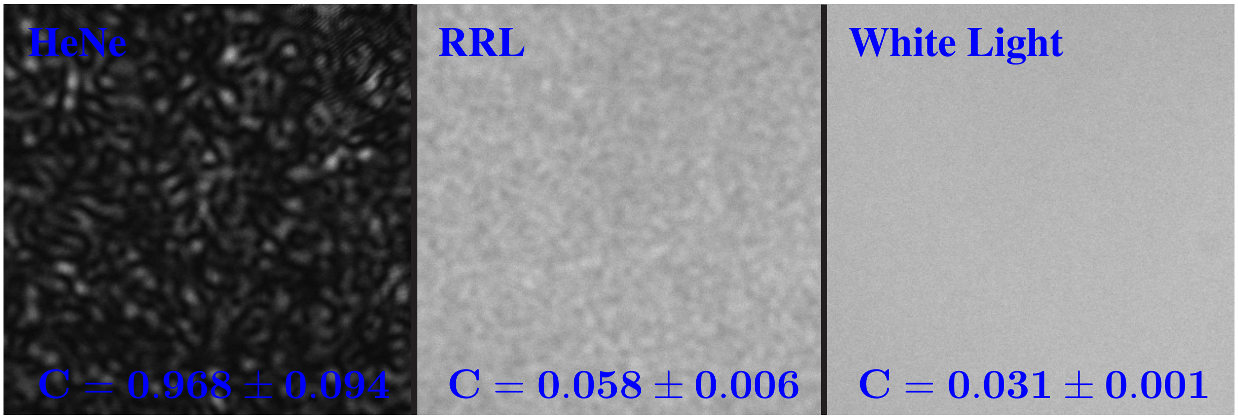

Figure 3. Speckle contrast of random Raman lasing. The speckle contrast measurements for a helium-neon laser and a halogen white light are shown for comparison and to illustrate the dynamic range of the measurement technique. This predicts 300 independent lasing modes present in the random Raman laser emission.

coefficient separately. These parameters are further defined and the simulations discussed in greater detail in a previous paper.[13] It is important to point out that the Monte Carlo simulations should account for all the relevant physical dynamics with the exception of any which arise due to the wave nature of light.

The steady-state ab initio laser theory (SALT) was designed to provide an exact treatment of the open cavity boundaries and spatially varying gain competition.[16–19] It yields a set of coupled differential equations in the frequency domain which can be efficiently solved numerically. Unlike previous Raman laser theories, this new theory self-consistently determines both the number of lasing modes and their frequencies, thus naturally accounts for both multi-mode operation and line-pulling effects between the natural Stokes frequency and the passive cavity resonances of the laser cavity. Using this new theory, we can directly compare the multi-mode proclivities of Raman gain media against traditional gain media for the same exact cavity holding all other relevant parameters, such as the gain width, equal. The results of such a comparison can be seen in Figs. 2A and 2C, which show the modal intensities as a function of pump strength in a random, 1-dimensional cavity for both a Raman and atomic gain medium respectively. Here, we see that the Raman gain medium usually only has a single mode active for any given pump power, in contrast with the atomic gain medium which is able to achieve multi-mode operation for most input pump strengths. We can understand this through looking at the associated lasing mode profiles shown in Figs. 2B and 2D. For the Raman laser, as the pump power is increased, the high Q lasing mode deep within the random cavity is eventually out-competed by the second lasing mode, which is lower Q, but exists closer to the edge of the cavity, and as such depletes the pump photons to the point where it drives the first lasing mode below threshold, turning it off. While the modes in the atomic medium do compete for gain with one another, this competition is through spatial hole-burning, rather than pump field depletion, which is an effect specific to χ^3 type gain media and parametric processes. These early results indicate that within a single cavity, there can be at most a few simultaneous steady-state Raman lasing modes, many fewer than are possible with an atomic gain medium. Thus, to increase the multi-mode behaviour of Raman lasers, one must simultaneously pump a large number of effective cavities, in which each cavity exists only a few highly localized modes, which do not compete for gain with other neighbouring effective cavities.

To assess the number of lasing modes present in the random Raman laser, the speckle contrast was measured.[20] The speckle contrast is defined to be

$$C = \frac{\sigma}{\langle I \rangle} = \frac{1}{\sqrt{m}} \tag{1}$$

where $\langle I \rangle$ is the average value of the intensity, σ is the standard deviation, and m is the number of independent modes present. Using these definitions, we find that there are 300 independent lasing modes present in the emission. This well exceeds the predictions of only a few simultaneous lasing modes in the theory presented above.

To measure the speckle contrast, the light is coupled into a 2 m long 600 μm fiber. The light from the fiber is left uncollimated and was allowed to overfill the array of a 16-bit CCD (Orca-ER; Hamamatsu). The CCD was placed far enough from the fiber that the speckle grain size is significantly larger than the pixel size of the CCD. To demonstrate the dynamic range of the measurement technique, a helium neon laser was used as a highly coherent source, and a halogen light as a low-coherence source. The resulting images and speckle contrast are shown in Fig. 3. The speckle contrast was measured by selecting a 400x400 region of the CCD that was clear of debris. That region was further subdivided into 25 80x80 sub-regions. The speckle contrast of each sub region was computed independently by first subtracting a pedestal, to reduce the effect of the CCD dark counts on the calculation, and then computing the mean and standard deviation.[21,22] This helps minimize any effects due to non-uniform illumination of the array. Multiple images were taken with each light source, and those used in the calculations for the random Raman lasing and halogen light source were selected by rejecting any images with saturated pixels and requiring the average counts per pixel to be greater than 20,000. This minimizes the effect of noise due to the CCD while ensuring that saturation is not occurring. For the Helium-Neon (HeNe) images, the requirement on average pixel count was not enforced due to the high contrast, but the laser power was adjusted to maximize the dynamic range of the detector without saturation. At the end, 21 images were used for the HeNe calculations, 28 images for the random Raman lasing emission, and 101 images for the white light. All of the sub regions from all of the images were first pooled together into a single data set and then averaged. The standard deviation of these calculations were used as the error.

3. CONCLUSION

There are several key measurements where random Raman lasing deviates from classical, particle-like, diffusion predictions, and while none of these measurements by itself constitutes the proverbial "smoking-gun" of Anderson localization, Anderson localization appears to be the only single effect that is known which is consistent with all of these measurements simultaneously. For this reason, we conclude that it appears highly likely that Anderson localization plays a key role in the dynamics of random Raman lasing, making it a fascinating platform to test our theoretical understanding of wave-like diffusion in non-linear systems.

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