Fluctuation and Noise Letters
Vol. 6, No. 1 (2006) L95–L101

World Scientific
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LÉVY STATISTICAL FLUCTUATIONS FROM A RANDOM AMPLIFYING MEDIUM

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Received 31 August 2005
Revised 9 January 2006
Accepted 10 January 2006
Communicated by Stuart Tessmer

We report our studies of emission from a dye-scatterer system, commonly known as random amplifying medium (RAM). It is found to exhibit non-Gaussian statistics of emission intensity over the ensemble of random realizations. The amplification is dominated by certain improbable events that are "larger than rare", which give the intensity statistics a Lévy-like fat tail. This, to the best of our knowledge, provides the first experimental realization of the Lévy statistics in the optics of a random amplifying medium, and the analysis thereof. Notably, the Lévy exponent is continuously tunable parametrically.

Keywords: Lévy statistics; random amplifying medium; multiple scattering.

In this Letter, we examine the statistics of fluctuations in the intensity of emission from a random amplifying medium (RAM) over different realizations of the randomness. We expect that the large gain of the amplifying medium and the random photon paths of varying lengths, will combine to give rise to some interesting features, such as non-selfaveraging of the scattering effects.[1] Indeed, we find, both experimentally and theoretically, that for a range of parameters of the RAM, the intensity fluctuations are non-Gaussian and, in fact, show Lévy statistics with tunable exponent. Beginning with a brief introduction to the RAM, we proceed to describe our experiments. We then present a theoretical treatment of the fluctuations in the intensity of emission from a RAM, and following this, analyze the experimental results in the light of the theoretical findings.

[1]See, e.g. P. A. Mello and N. Kumar, *Quantum transport in mesoscopic systems: Complexity and statistical fluctuations*, Oxford University Press (London,2004); Note that the non-selfaveraging is usually invoked in the context of wave-scattering in, and transmission through a random medium where it is due to wave interference. In the present context of classical diffusion, non-selfaveraging of emission arises from the high gain of the random amplifying medium that accentuates the effect of the rare long paths.

A RAM is typically a dye-scatterer system — that is, a solution of a laser dye (e.g. Rhodamine in methanol) in which point-like scatterers (e.g. polystyrene microspheres) are randomly suspended. We recall that, unlike conventional lasers which demand clean homogeneous amplifying media placed in a resonant cavity, usually formed by mirrors, with minimal scattering, as these result in losses that are detrimental to lasing, a RAM is a non-resonant system with distributed feedback that combines multiple scattering with high gain to give rise to lasing [1–3]. The dye-scatterer system is pumped optically to create a population inversion in the dye. Any spontaneously emitted photon is amplified as it travels through the pumped region. Due to the randomly placed scatterers, the photon undergoes multiple scattering and its pathlength within the active medium, before it finally exits the dye-scatterer system, is increased. Consequently, the amplification of the light is enhanced resulting in lasing when the gain exceeds the loss.

In a RAM light is both multiply scattered and amplified. The characteristic length scales that describe the scattering process are the scattering mean free path (l_s) and the transport mean free path (l_t). The former is defined as the average distance between two successive scattering events and is given as : $l_s = 1/n\sigma$, where, n is the number density of scatterers and σ is the scattering cross-section of an individual scatterer, which depends on the size of the scatterer, incident wavelength and refractive index mismatch between the scatterer and its surroundings. The scattering mean free path being inadequate to account for the anisotropy in scattering, the transport mean free path (l_t) is defined as the average distance that the light travels before its direction of propagation is randomized and is given as $l_t = l_s/(1-g)$, where, the anisotropy parameter is $g =< cos\theta >$, θ being the scattering angle. The length scales relevant to the amplification within a RAM, are the gain length (l_g) and the amplification length (l_{amp}). In a gain medium without scattering, the intensity I increases exponentially, as $I(l) = I_o e^{l/l_g}$. In a RAM, due to multiple scattering and the consequent random paths of the photons, the actual length of travel of a photon is much greater than the distance between the beginning and the ending points giving rise to the concept of l_{amp}, the average distance between the begin and the end points for which the actual path length of the photon is l_g. This is given by $l_{amp} = \sqrt{l_s l_g/3}$.

In a RAM, the gain due to amplification competes with the losses due to the escape of photons from the bounding surface of the amplifying medium and self-absorption by the dye. Lasing is experimentally observed as a drastic spectral narrowing (gain narrowing) of the emission from a few tens of nanometers to a few nanometers and occurs above a well-defined threshold of pulsed-pump power. This is illustrated in Figs. 1(a) and (b) which give the emission spectra obtained by us from a sample of Rhodamine 6G (R6G) in ethanol (10^{-2}M) containing polystyrene microspheres (mean diameter = 0.30 μm, number density = 6.278×10^{11}/cc) at pump energies below and above threshold ($\sim$ 15 μJ and $\sim$ 50 μJ respectively), where the linewidths are $\sim$40nm and $\sim$9nm respectively. Typically, the threshold of the pump-power for linewidth collapse is almost two orders of magnitude smaller in the system containing scatterers than the one without [3, 4] — the lowering of threshold being greater the larger the number density of scatterers.

At pump powers much above threshold, the nature of the spectrum changes drastically; instead of a smooth peak with a width of few nanometers, several ex-

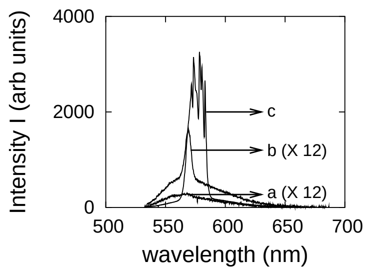

Fig. 1. Emission spectra from R6G (10^{-2}M) dye in ethanol containing polystyrene microspheres (mean diameter = 0.30 μm, number density = 6.278×10^{11}/cc), as a function of pump energy. (a) $\sim$ 15 μJ (b) $\sim$ 50 μJ (c) $\sim$ 1 mJ.

tremely narrow spikes ($\sim$ 0.2nm) appear in the spectrum [5–7]. This is illustrated for our system under study, in Fig. 1(c), at a high pump energy ($\sim$ 1 mJ). Our aim in the present study is to examine the statistics of the pulse-to-pulse fluctuations in the emission intensity from the RAM. This is studied at a particular wavelength, as function of various parameters, such as pump power (gain), characterising the system.

Before we move on to the experiment and the theoretical derivation, we briefly recall the distinguishing features of the Gaussian and the Lévy statistics relevant to our work. In the normal random walk, the total displacement (X) has a Gaussian distribution (with finite mean and variance). However, Lévy statistics [8] arises when the variance diverges with an asymptotic probability distribution P(X) given by P(X) $\sim |X|^{-(1+\nu)}$ with the exponent $\nu < 2$ ($\nu \geq 2$ corresponds to the Gaussian case). Physically, the Lévy statistics is well known for random variables having orders-of-magnitude larger values but with correspondingly orders-of-magnitude smaller probabilities for their occurrence. For these "larger-than-rare" events, the variance may diverge, with a single large event dominating the sum of a large number of such random events, unlike the normal Gaussian case. Many physical examples of the Lévy statistics are known, e.g. strange kinetics [9], anomalous diffusion in living polymers [10], subrecoil laser-cooling [11], rotating fluid flow [12] and interstellar scintillations [13]. Ours, is to the best of our knowledge, the first reported observation of Lévy statistical fluctuations of emission from a RAM.

In the present work we experimentally demonstrate the transition from Gaussian to Lévy statistics in the fluctuations of emission from the RAM over different realizations. These random realizations can originate variously in our experiments For example, there is randomness inherent in the diffusive motion of the photon through the RAM. Further, the configuration of the scatterers may also be changing constantly (because the scatterers are suspended in liquid dye solution) during the interval between successive pump pulses (shots). This constitutes effectively a literal sample-to-sample fluctuation.

The experimental studies were conducted on the dye-scatterer systems consisting of R6G dye (5×10^{-3} M and 10^{-2} M) in ethanol with sub-micron (TiO_2 or polystyrene microspheres) dispersed in them. The system, contained in a glass cuvette of size 1cm $\times$ 1cm $\times$ 5cm was irradiated by 35ps pulse at 532nm from a frequency doubled Nd:YAG laser. The incident pulse was split into two by a beam-

beamsplitter (R/T = 50/50). While the transmitted beam was focussed to a spot of diameter ~ 1.2 mm before it was incident on the sample, the reflected part was used to monitor the energy of the pump pulse by the energy meter (Laser Probe Inc., Rj-7620). The pump energy was maintained constant throughout an experimental run. The emission was collected and focussed onto an optical fibre and the spectrum recorded on a PC based fibre optic spectrometer (Ocean Optics S2000). In order to obtain good statistics for pulse-to-pulse fluctuation of emission intensity, five hundred such single-shot spectra were recorded. The pulse-to-pulse fluctuations in emission intensity were studied over a wide range of pump energy, dye concentration, scatterer density and type of scatterers. Typical spectra are as shown in Figs. 1(a), (b) and (c). From the five hundred spectra obtained for each sample, for a given set of parameters e.g. pump power, intensity histograms were constructed for a chosen wavelength, by plotting the number of times an intensity value was obtained (normalized to the total number of spectra), as function of the intensity.

Figure 2(a) gives the histogram observed from spectra for an R6G-TiO_2 sample, recorded at near-threshold pumping (~ 90 μJ), where the emission has a spectral width of $\sim$10 nm. The histogram is Gaussian. Figure 2(b) gives the histogram when the pump power was increased to 3 mJ (and the spectra showed several spikes). At high emission intensities, the histogram deviates from Gaussian and shows a fat tail. This is a signature of Lévy statistics, and is to be expected from the following theoretical considerations.

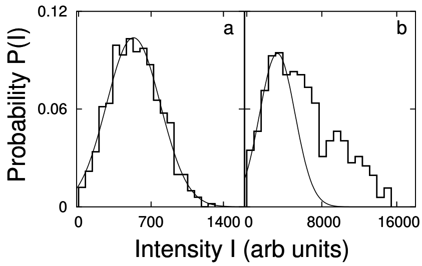

Fig. 2. Histograms as a function of pump energy for R6G (10^{-2}M) in ethanol containing TiO_2 microspheres (mean diameter = 0.36 μm, number density $\sim 10^{10}$/cc). (a) ~ 90 μJ, showing Gaussian statistics (b) ~ 3 mJ, showing Lévy-like fat tail.

Consider a weakly scattering (diffusive) RAM composed of random point-like scatterers dispersed in an amplifying continuum. A spontaneously emitted photon diffuses with diffusion constant $D = cl_t/3$, (c is the speed of light in the medium) getting amplified as it propagates with gain $g = e^{ct/l_g}$, and finally exiting after time t. We assume, for simplicity, a spherical RAM (radius 'a'), illuminated uniformly by a short pump-pulse at time $t = 0$. The rate of emission is the probability of escape of a photon from a surface ($r = a$) per unit time, with the photons spontaneously created at time $t = 0$ within the RAM at positions r, and is given by

$$p_I(t) = -\frac{\partial}{\partial t}\int_0^a \rho(\mathbf{r}, t) d^3 r\,, \qquad (1)$$

where, $\rho(\mathbf{r}, t)$ is the probability density of the diffusing photon, emitted initially anywhere within the sample with a uniform initial probability density ρ_o. This requires the solution for the diffusion equation

$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} = D\nabla^2 \rho(\mathbf{r},t) \tag{2}$$

which, on solving gives

$$\rho(\mathbf{r},t) = \rho_o \sum_{m=1}^{\infty} \frac{2a}{\pi m}(-1)^{m+1} \cdot \frac{sin(\frac{\pi m r}{a})}{r} e^{-\frac{\pi^2 m^2}{a^2} Dt} \tag{3}$$

giving straightforwardly

$$p_I(t) = \rho_o \sum_{m=1}^{\infty} 8aD\pi \; e^{-\frac{\pi^2 m^2}{a^2} Dt} . \tag{4}$$

With a change of variable, the probability distribution for the gain $p_g(g)$ is obtained as

$$p_g(g) = \sum_{m=1}^{\infty} \frac{\rho_o 8aD\pi l_g}{c} \frac{1}{g^{1+\alpha_m}} \equiv \sum_{m=1}^{\infty} \frac{8\rho_o a\pi l_t l_g}{3} \frac{1}{g^{1+\alpha_m}} \tag{5}$$

with $\alpha_m = m^2(\pi^2 l_t l_g/3a^2) \equiv$ the mth Lévy exponent. Thus, with increasing pumping or increasing dye concentration (decreasing l_g), the exponent α_m decreases, the tail becomes fatter, and the variance of g diverges for $\alpha_m < 2$, that happens first for $m = 1$, i.e., for $(\pi^2 l_t l_g/3a^2) < 2$. This leads to the controlled crossover from a finite variance (Gaussian) to a divergent variance (Lévy) limit. Similar control can also be achieved by increasing the number density of scatterers or by enhancing the refractive index contrast between the active bulk and the passive scatterers (decreasing l_t). Below we discuss the cases for the different control parameters realized experimentally.

The two histograms of Fig. 2 are now readily understood as corresponding respectively to the low and the high gain cases. Similar effect should be, and is indeed observed by increasing the dye concentration which reduces l_g. Figures 3(a) and (b) show the histograms for an R6G-TiO_2 system at two different dye concentrations (5×10^{-3}M and 10^{-2}M respectively), all other parameters like pump energy, scatterer density kept the same. Clearly, 10^{-2}M dye solution shows more pronounced Lévy feature than 5×10^{-3}M dye solution.

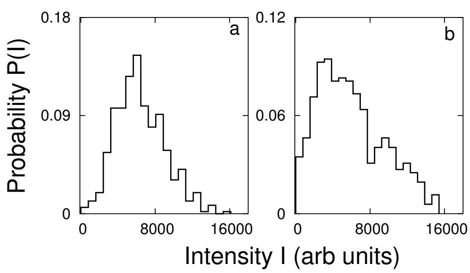

Fig. 3. Histograms for R6G-TiO_2 system (scatterer density $\sim 10^{10}$/cc, pump energy ~ 3 mJ) with dye concentration (a) 5×10^{-3}M (b) 10^{-2}M.

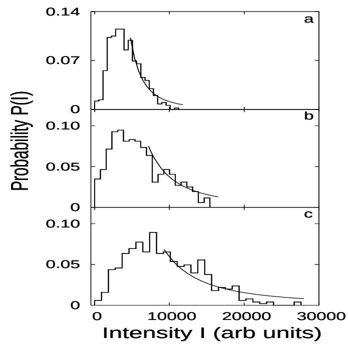

Fig. 4. Histograms for R6G(10^{-2}M)-TiO_2 system (pump energy $\sim$ 3 mJ) for scatterer number density (a) $\sim 10^9$/cc (b) $\sim 10^{10}$/cc (c) $\sim 2 \times 10^{10}$/cc.

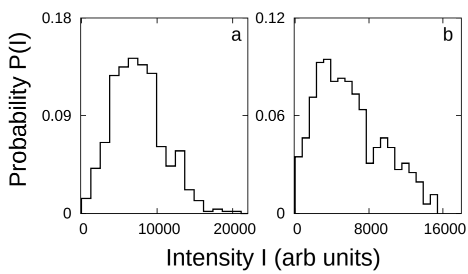

Fig. 5. Histograms for R6G (10^{-2}M) dye in ethanol (pump energy $\sim$ 3 mJ) containing (a) polystyrene microspheres (mean diameter = 0.30 μm, number density = 6.278×10^{11}/cc) (b) TiO_2 (mean diameter = 0.36 μm, number density $\sim 10^{10}$/cc).

The expression for α shows that the transition from Gaussian to Lévy may also be obtained by altering l_t. Figures 4(a), (b) and (c) show the histograms for R6G (10^{-2}M) in ethanol with suspension of TiO_2 microspheres with number density of $\sim 10^9$/cc, $\sim 10^{10}$/cc and $\sim 2 \times 10^{10}$/cc respectively. Clearly for low scatterer density the statistics is almost Gaussian (Fig. 4(a)). As the scatterer number density is increased, a noticeable Lévy-like tail appears (Fig. 4(b)), which when fit to the power law $\sim \mathrm{g}^{-1-\alpha}$ gives the Lévy exponent (α) of 1.13. On further increasing the scatterer concentration, the tail becomes quite pronounced with a Lévy exponent (α) of 0.94 (Fig. 4(c)), demonstrating the tunability of the Lévy exponent. In addition, more pronounced Lévy features were observed with a R6G-TiO_2 system (Fig. 5(a)), than in a R6G-polystyrene system (Fig. 5(b)), despite the former containing fewer scatterers. This is because, TiO_2 has a refractive index $\sim$ 2.7, while polystyrene $\sim$ 1.59, the former provides a higher refractive index mismatch with the surrounding solvent (ethanol, refractive index $\sim$ 1.36) resulting in enhanced scattering (smaller l_t) and amplification. Thus, the transition from Gaussian to Lévy has been engineered by four different means, two of which alter l_g and two l_t.

The occurrence of Lévy statistics in the dye-scatterer system can be understood physically in terms of the distribution of the path lengths of photons in the gain medium. In the case of very dense scattering media, some of the photons undergo very many scatterings, so that their cumulative pathlengths within the medium is extremely long. The gain in the system makes the associated amplification considerable, and these photons then dominate the emission, leading to enhanced intensity whenever such long paths occur. Their occurrence is rare, and is noticeable only in systems with large scattering or high gain. This is consistent with the recent numerical simulations [7].

In conclusion, we have shown theoretically that the emission intensity fluctuations from a random amplifying medium can exhibit Gaussian or Lévy statistics, depending on the scattering and gain parameters. A transition from one regime to the other may be brought about, and the Lévy exponent tuned, by altering these two parameters. We have experimentally demonstrated this in dye-scatterer RAMs.

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