Lévy statistics of emission from a novel random amplifying medium: an optical realization of the Arrhenius cascade

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Received January 25, 2006; revised March 15, 2006; accepted March 15, 2006; posted March 30, 2006 (Doc. ID 67460)

We report the observation of Lévy-like statistical configuration-to-configuration fluctuations in the intensity of emission from a novel system, the fiber-random amplifying medium, where active fiber segments are embedded randomly in a bulk of pointlike passive scatterers. Some rare configurations of fibers provide long, guided amplifying paths for the photons, leading to high jumps in the intensity, and thus to Lévy statistics. This system provides an optical realization of the Arrhenius cascade. © 2006 Optical Society of America

OCIS codes: 290.4210, 290.5850, 000.5490.

We recently reported[1] Lévy-like statistical fluctuations in the emission intensity from a dye-scatterer random amplifying medium[2–6] (RAM) over its random configurational realizations.[7] The large jumps in intensity resulted from rare events where a photon traversed a long path in the gain medium owing to multiple scattering and was significantly amplified. Lévy statistics[8–13] occurs in systems where certain improbable events that are larger than rare dominate. The present work was undertaken to establish the crucial role of the rare long amplifying paths in causing Lévy fluctuations in a RAM. Experiments were carried out in a novel system, a fiber-random amplifying medium (F-RAM), which is an aggregate of segments of active fiber embedded randomly in a passive scattering bulk. This tailored system permits intentional introduction of long paths with the desired probability of occurrence. The mean length of the fiber segments was much greater than the mean free path of light in the passive scattering background, making the probability of prompt escape from the sample much higher. Thus the long photon paths in the F-RAM are even rarer than those in a RAM. The high gain in the fiber, however, makes it possible to observe these very rare events as random large jumps in the emission intensity.

The F-RAM is based on the classical Arrhenius cascade model[14] that examines the total time $t=\Sigma_{i=1}^{n}t_i$ spent by a particle in going down an inclined washboard of n potential wells. The probability density $p(U_i)$ for the well depth (U_i) is $p(U_i)=(1/U_o)e^{-(U_i/U_o)}$ and activated trapping time $t_i=t_o e^{(U_i/kT)}\equiv t_i(U_i)$. The two may be combined by using the law of probability, $p(t_i)\mathrm{d}t_i=p(U_i)\mathrm{d}U_i$, to yield an inverse power-law probability density for the trapping time, $p(t_i)\sim t_i^{-(1+\alpha)}$ with $\alpha=kT/U_o$. Thus, for $\alpha\geqslant 2$, the second moment is finite, and the total time $(t/n^{1/2})$ probability density tends to the Gaussian limit—the classical central limit theorem—as $n\to\infty$. For $0<\alpha<2$, however, the second moment of the individual trapping time diverges, and the corresponding total time $(t/n^{1/\alpha})$ tends to the Lévy distribution with an inverse power-law probability density $\sim t^{-(1+\alpha)}$ asymptotically (the generalized central limit theorem, α being the Lévy exponent). Thus, we have the Gaussian statistics at high temperatures and the Lévy statistics at low temperatures for the cascade. In our optical analogy, we create an F-RAM system from segments of amplifying fibers and study the cumulative gain of a photon diffusing through the F-RAM. The lengths of the segments are tailored to follow an exponential distribution $p(l_i)=(1/l_a)e^{-(l_i/l_a)}$, where l_a is the mean length of the fiber. Photons originating within the sample travel through the fibers of random lengths and finally exit with a total gain $g=e^{(l/l_g)}$, where l is the total fiber length traversed and l_g is the gain length of the active fiber. Thus, in an F-RAM, l_a and l_g are analogous to U_o and T, respectively, of the Arrhenius cascade. This suggests that the Lévy exponent may be tuned by altering the gain or the length distribution of the fiber.

For a diffusive dye-scatterer RAM, the probability density for the total gain $p(g)$ acquired by a spontaneously emitted photon as a result of multiple scattering in the amplifying medium is given[1] by

$$p(g)=\sum_{m=1}^{\infty}\frac{8\rho_o a\pi l_t l_g}{3}\frac{1}{g^{1+\alpha_m}}. \tag{1}$$

While this analytical expression does not literally simulate the discrete Arrhenius cascade, it does show the qualitative feature of large but rare fluctuations, permitting Lévy-like statistics. For large g, the expression for $p(g)$ is dominated by the smallest exponent α_1 that can be identified with the Lévy exponent (α). For a sum of independent random variables, the only limiting distributions admissible are the Gaussian and the Lévy, according to whether the variables have finite or infinite variances.[15] Our analytical result [Eq. (1)] for the RAM is, strictly speaking, neither of these possibly because the total amplified photon output is a combination of additive and multiplicative processes taking place in the RAM.

We first generalize the analytic treatment of the conventional RAM for an F-RAM through an effective-medium theory. Here, the F-RAM is considered a composite medium with a volume fraction v_a of the active medium embedded in a passive, diffusive medium of volume fraction v_p with transport mean

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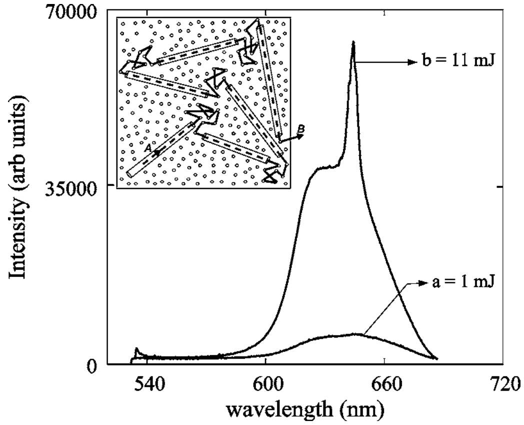

Fig. 1. Spectra from an F-RAM at (a) below and (b) above threshold pumping. Inset, schematic of an F-RAM, a typical photon path ($A \rightarrow B$), with amplifying regions shown by dashed lines.

free path l_p. The amplifying medium is a random aggregate of active fiber pieces of radius a and mean length l_a (mean volume $\pi a^2 l_a$). As the fiber provides wave guidance over its length (few millimeters) and is much greater than the transport mean free path in the passive bulk (a few micrometers), the direction of propagation of a photon upon exiting a fiber is rapidly randomized. Thus the mean free path of the photons in the *active* medium is l_a itself. We now combine these two random components into the simplest effective medium, using the Matthiessen rule.[16] Thus, the photons diffuse in the F-RAM with a diffusion constant $D=cl_{\mathrm{eff}}/3$, where $1/l_{\mathrm{eff}}=v_a/l_a+v_p/l_p$, and undergo amplification $e^{v_a ct/l_g}$ in time t. Proceeding as before,[1] we obtain

$$p(g) = \sum_{m=1}^{\infty} \frac{8\rho_o a \pi l_{\mathrm{eff}} l_g}{3v_a} \frac{1}{g^{1+\alpha_m}}, \qquad (2)$$

with $\alpha_m = m^2\pi^2 l_{\mathrm{eff}} l_g / 3a^2 v_a$. This suggests a Lévy-like fat tail for $\alpha_1 < 2$ and a finite second moment akin to Gaussian behavior for $\alpha_1 \geqslant 2$. The Lévy exponent is now tunable by gain and also by l_{eff}, which depends on l_a, l_p, v_a, and v_p. These features are confirmed *a posteriori* through our experiments discussed below.

Our experimental studies were conducted on an F-RAM that contained a random aggregation of segments of dye-doped amplifying fibers (Bicron, fluorescent red) embedded in a passive medium of granular starch (inset of Fig. 1). These plastic fibers fluoresce in the orange-red when pumped by green light, which can enter the fibers through their cylindrical surfaces anywhere along their lengths. The emitted light is, however, guided mainly along the length of the fiber, and it emerges from either end amplified by a factor that increases exponentially with its path length in the fiber. The random aggregation of the amplifying fibers itself provides some scattering, which is enhanced by the addition of passive scatterers. While the usual RAM is a bulk (3-dimensional) active medium with pointlike (0-dimensional) scatterers embedded in it, an F-RAM has a passive, scattering bulk with active (1-dimensional) fibers embedded in it. Thus in an F-RAM the diffusion proceeds via random scattering in the passive bulk and wave guidance, through the randomly embedded active fiber segments.

Our sample had 500 segments of amplifying fibers, with lengths (l_i) in the range 1–20 mm, following the exponential distribution $p(l_i)$ with l_a=5 mm. Granular starch ($\sim$1 mm^3), with negligible absorption at the wavelengths of interest, was added to provide the random passive scattering background. The F-RAM was contained in a glass cuvette of size 3 cm $\times$ 3 cm $\times$ 6 cm and pumped optically by 10 ns pulses at 532 nm from a frequency-doubled Nd:YAG laser. The emission from the system was collected in a direction transverse to the pump beam, and the spectrum was recorded with a PC-based fiber-optic spectrometer. The F-RAM showed broad emission (width $\sim$30 nm) typical of dyes at low pump energies and exhibited gain narrowing (width $<$5 nm) with the intensity peaking at $\sim$640 nm at above threshold. (3–5 mJ) (Fig. 1).

As an F-RAM is a random aggregation of segments of active fiber, many configurations are possible for the same macroscopic composition of the F-RAM. Our experiment was aimed at studying the variation in emission intensity over these random configurations, and, therefore, the sample was stirred to change the configuration. The corresponding spectra for 420 dif-

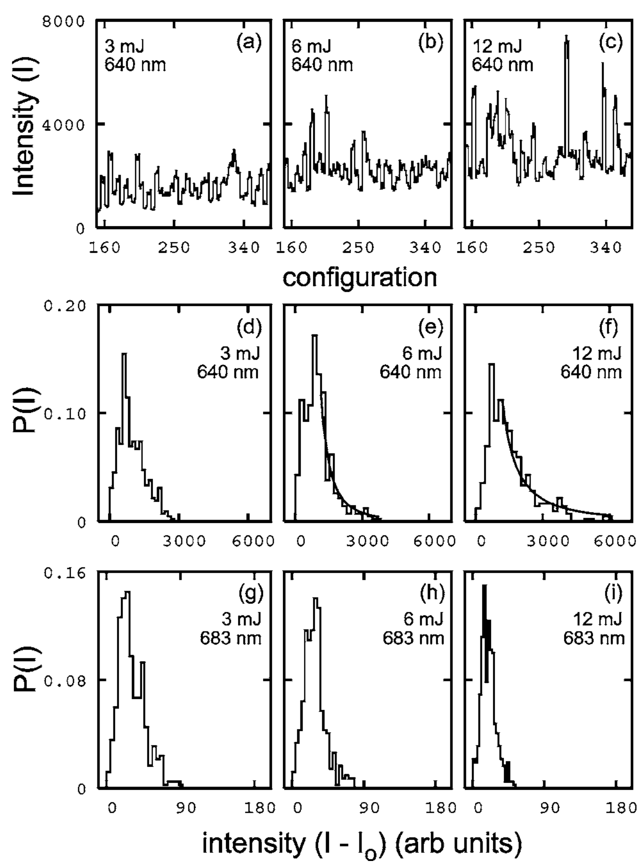

Fig. 2. (a)–(c) Intensity versus configuration, (d)–(f) corresponding histograms fitted to the power law $(I-I_o)^{-(1+\alpha)}$: I_o =619, 1267, and 1299 for (d), (e), and (f), respectively; (g)–(i) histograms at λ_{op}: I_o=17, 24, and 12 for (g), (h), and (i), respectively.

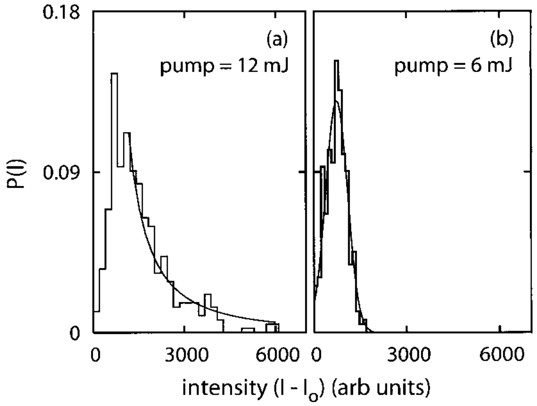

Fig. 3. Histogram at λ_p for (a) F-RAM with power-law fit, $\alpha = 0.82$ and (b) powdered-F-RAM and Gaussian fit.

ferent configurations of the F-RAM were recorded. Histograms were then constructed by plotting the number of times an intensity value was obtained (normalized to the total number of spectra) as a function of intensity. Note that our samples are large enough to ensure spatial ergodicity.

Figures 2(a)–2(c) give the intensity values at the peak emission wavelength (λ_p) as a function of configuration for the F-RAM at three pump energies (3, 6, 12 mJ), and Figs. 2(d)–2(f) give the corresponding histograms. Clearly, the configuration-to-configuration fluctuations are small at the subthreshold pump energy of 3 mJ [Fig. 2(a)], and the statistics is Gaussian [Fig. 2(d)]. Slightly above threshold (6 mJ), the intensity values show a few large jumps as the configuration is altered [Fig. 2(b)] and a tail develops in the corresponding histogram [Fig. 2(e)]. At a much higher pump energy (12 mJ), the intensity values show many large jumps [Fig. 2(c)] with a pronounced Lévy-like tail in the histogram [Fig. 2(f)].

A fit to the power law $p(g) \sim g^{-(1+\alpha)}$ gave exponents $\alpha = 1.77$ and $\alpha = 0.82$ at pump energies of 6 and 12 mJ, respectively. When the pumping is doubled, the exponent is found to be halved,[17] as expected from the expression for α_1 [Eq. (2)]. At off-peak wavelengths (λ_{op}), the configuration-to-configuration fluctuations remained small, and the histograms exhibited Gaussian statistics at all the three pump energies [Figs. 2(g)–2(i)].[18]

We now discuss the experimental results in light of our earlier analysis, considering first the case of above-the-threshold pumping. Typically, a photon traverses a large number of short steps as it is multiply scattered by the passive scatterers or is guided by very short active fiber pieces, resulting in configuration-to-configuration fluctuations in the emission intensity that are small and of nearly the same magnitude. Occasionally, however, a long segment of active fiber guides it over relatively large lengths and amplifies it considerably, causing a sudden large jump in intensity. This is similar to a Lévy flight, and we may aptly term such an F-RAM a Lévy laser. At subthreshold pumping, however, the gain is small, and the emission even from a long fiber is small, and hence the Gaussian statistics of the fluctuations.

To verify that the presence of the exponentially rare long paths was crucial to the Lévy statistical fluctuations, we made a powdered F-RAM by cryogenically crushing the active medium to nearly uniform submillimeter sizes ($l_a \ll l_g$) and adding it to passive granular starch, keeping v_a and v_p the same as in the F-RAM. The two systems differed only in that the F-RAM contained exponentially rare long pieces of active fiber, while the powered F-RAM had only short pieces. Spectra for 420 configurations of the powdered F-RAM were recorded, and a histogram for λ_p was constructed. The statistics was clearly Gaussian [Fig 3(b)]. In contrast, Lévy statistics was obtained for the F-RAM at much lower pump energies [Fig 3(a)], confirming the role of the rare long paths in leading to the Lévy-like fat tail.

To conclude, though the main emphasis in this work is on Lévy statistics with a tunable exponent in RAMs, Lévy flight is simulated by adding segments of active fiber to a passive scattering medium. This F-RAM also provides an optical realization of the Arrhenius cascade.

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