

Fluctuations in a Diffusive Medium with Gain

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We present a stochastic model for amplifying, diffusive media such as, for instance, random lasers. Starting from a simple random-walk model, we derive a stochastic partial differential equation for the energy field which contains a multiplicative random-advection term yielding intermittency and power-law distributions of the field itself. A dimensional analysis indicates that such features are more likely to be observed for small enough samples and in lower spatial dimensions.

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Wave transport in disordered media can be described as a multiple scattering process in which waves are randomly scattered by a large number of separate elements [1]. To the first approximation this gives rise to a diffusion process. A particularly interesting situation arises when gain is added to a random material. In optics this is realized, for instance, in the form of a suspension of microparticles with added laser dye or by grinding a laser crystal. If the total gain becomes larger than the losses, fluctuations grow and these systems exhibit a lasing threshold [2], yielding the so-called random laser (see, e.g., Refs. [3,4] and the references therein). The complexity generated by the interplay between gain and disorder leads to intriguing connections with other fundamental problems such as Anderson localization [5] or the physics of glasses [6]. In addition to their fundamental interest, random lasers are likely to have a technological impact as low-cost light sources.

Similar situations occur in other branches of physics such as neutron diffusion in fissile materials or stochastic wave growth [7] and acceleration of plasma particles [8]. As is known, the competition between growth and propagation or diffusion is also a basic mechanism of population dynamics and theoretical ecology [9].

Diffusive random lasing has been observed experimentally in various active random media, including powders, laser dye suspensions, and organic systems [10–12]. Theoretical descriptions often rely on the diffusive approximation either by reaction-diffusion type of equations [2,13] or, on a more microscopic level, by the master-equation approach [14]. Monte Carlo simulations of course play an important role [15,16].

One of the salient experimental features of random laser emission is its large statistical variability [17]. Indeed, already within the diffusive approximation, the addition of gain (and saturation) naturally generates fat-tailed distributions [18] that stem from rare long light paths [16]. This mechanism is of pure statistical origin and does not require localization or interference [17]. It was also proposed that such systems can exhibit Lévy-type statistics in the distribution of intensities [18–20], and crossovers among different statistics have been predicted [21].

Remarkably, those predictions were very recently confirmed experimentally [22] (see also Ref. [23]).

In this work we first present a simple stochastic model that can be analytically solved and that yields large statistical fluctuations. From it, we derive an equation for the energy field which contains a stochastic multiplicative term whose strength controls such anomalous fluctuations. The magnitude of this term introduces another scale in the problem which thus provides a criterion for the observability of such intermittency in the field distribution.

We choose to describe isotropic diffusion of light in terms of an ensemble of independent random walkers each carrying a given energy (number of photons). This may be visualized as an ensemble of “beams” propagating independently throughout the sample, each interacting with an underlying atomic population providing a gain mechanism via stimulated emission. This procedure of attaching an energy to a random walk of photons is a common practice in Monte Carlo simulations of absorption in complex materials such as, e.g., tissues [24]. It has also been employed previously for random lasers [16,21], under the implicit assumption that all the photons generated by amplification are diffused in the same direction at each scattering event.

Let us denote by x and E the walker’s position and energy. We discuss the one-dimensional case in which the walker resides on a finite interval, $0 \leq x \leq L$. The dynamics is formulated as follows. A new walker is generated at a random position by a spontaneous emission event with a rate γ and initial energy $E = \varepsilon$. In terms of the underlying active media, γ denotes the spontaneous emission rate of the single atom. The walker position x changes to $x \pm a$ according to a standard random-walk rule on a lattice with spacing a . At the same time, the walker energy E may increase by one unit due to the process of stimulated emission, $E \rightarrow E + \varepsilon$, with a rate $\Gamma(E)$. The simplest choice would be $\Gamma(E) = \gamma E$ or, to mimic saturation effects, we may consider a gain of the form $\Gamma(E) = \gamma E / (1 + E/E_s)$.

The probability $P_i(n)$ for the walker to be at $x = ia$ having an energy $E = n\varepsilon$ evolves according to a master

equation that can be solved by standard methods [25]. Here, to simplify further, we work directly in the continuum limit and treat x and E as continuous variables, obeying the Langevin equations [25]

$$\dot{x} = \sqrt{2D}\xi, \quad \dot{E} = \Gamma(E) + \sqrt{\Gamma(E)}\eta, \quad (1)$$

where ξ , η are δ -correlated Gaussian variables with zero average and $\langle \xi^2 \rangle = \langle \eta^2 \rangle = 1$ (from now on $\langle \cdot \cdot \cdot \rangle$ denotes an average over realizations of the process). To keep things as simple as possible, bulk absorption or the possibility that diffusion is affected by the energy is neglected from scratch but can be easily included.

To demonstrate that this dynamics naturally generate power-law distributions of energies, we solve the associated Fokker-Planck equation [Itô interpretation of Eq. (1)]

$$\dot{P} = D \frac{\partial^2 P}{\partial x^2} - \frac{\partial}{\partial E} \left(\Gamma P - \frac{1}{2} \frac{\partial \Gamma P}{\partial E} \right), \quad (2)$$

where $P(x, E, t)$ is the probability of finding a walker with energy E at x . Four boundary conditions are necessary on the contour of the domain $[0, L] \times [1, \infty)$. To account for complete absorption at the boundaries we let $P(0, E, t) = P(L, E, t) = 0$. Moreover, $P(x, 1, t) \equiv f(x, t)$ is determined as a solution of

$$\dot{f} = Df'' - \gamma f + \gamma, \quad (3)$$

where we approximated $\Gamma(1) \approx \gamma$. This condition may appear somehow unusual and is justified as follows: walkers with unit energy increase at rate γ and are free to diffuse without increasing their energy. Their number diminishes by a term $-\gamma f$ because they gain energy by amplification. Note that Eq. (3) can be derived exactly from the underlying master equation for the discrete variables [25].

The stationary solution of Eq. (2) can be found by separation of variables $P(x, E) = Q(x)W(E)$, yielding $Q \sim \sin(kx)$ with $k = m\pi/L$ (m integer) being the separation constant (the wave number) that somehow couples diffusion and gain. The physical origin of such a coupling is that E depends on the actual path length spent by the walker within the sample: longer paths acquire a larger energy and the high- E statistics ultimately depend on this mechanism. The equations for W may be integrated exactly, but for our purposes it suffices to solve an approximated form where the energy-diffusion term in Eq. (2) is neglected, $W \approx \exp[-Dk^2 \int^E \Gamma^{-1}(E') dE'] / \Gamma(E)$. Thus, for the linear gain W has a power-law tail while for the saturating case

$$W(E) \propto \frac{\exp(-Dk^2 E / \gamma E_s)}{E^{1+Dk^2/\gamma}} \quad (4)$$

(we have also assumed $E_s \gg E \gg 1$). The general solution is a sum over the allowed k 's. As a further approximation, we consider only the first Fourier mode $k = \pi/L$. This is mathematically justified noting that Q is fixed by

the stationary solution of Eq. (3) which for $f(0) = f(L) = 0$ is ($\beta \equiv \sqrt{\gamma/D}$)

$$f(x) = 1 + \frac{\sinh(\beta(x-L)) - \sinh(\beta x)}{\sinh(\beta L)}. \quad (5)$$

In the critical region (to be defined below), $\beta \sim \pi/L$ so that f (and thus Q) is indeed very close to the shape of the first Fourier mode itself. The result is that the distribution of energies displays a parameter-dependent power law

$$P(x, E) \propto \sin\left(\frac{\pi x}{L}\right) \frac{\exp(-\alpha E/E_s)}{E^{1+\alpha}}, \quad (6)$$

where $\gamma_c \equiv D(\pi/L)^2$ and $\alpha \equiv \gamma_c/\gamma$, which is exponentially cutoff at $E \sim E_s$. The origin of the power law (6) is traced back to very long paths which, although exponentially rare, acquire an exponentially large energy while diffusing throughout the sample [18,21]. Note that $\gamma = \gamma_c$ correspond to the case of a Cauchy-like tail $\alpha = 1$, $P(x, E) \propto E^{-2}$, yielding a diverging average value of the energy for $E_s \rightarrow \infty$. It is thus natural to identify this as the ‘‘laser threshold’’ for the model: it will be shown below that this coincides with the usual criterion of gain overcoming the losses.

Thus far we have described the system in terms of a single walker property. Consider a population of M walkers, and denote by x_i and E_i their position and energies, each obeying the equation of motion (1). We thus introduce the energy density field

$$\phi(x, t) = \sum_i^{M(t)} E_i \delta(x - x_i(t)). \quad (7)$$

The number $M(t)$ fluctuates in time due to the fact that walkers are created (at a rate γL) and absorbed at the boundaries [at a rate $D(\pi/L)^2 M$], so that $M \sim \gamma L^3/D$. For $\gamma \sim \gamma_c \sim D/L^2$ (which is the regime of interest here) $M/L \sim 1$; i.e., there are a few walkers per unit length and fluctuations must be very relevant there. The problem can thus be recast [26] in terms of a Langevin equation for ϕ [25]:

$$\frac{\partial \phi}{\partial t} = D \frac{\partial^2 \phi}{\partial x^2} + \Gamma(\phi) + \frac{\partial}{\partial x} (v\phi) + s. \quad (8)$$

As done above, we neglected the $\sqrt{\Gamma}$ term in Eq. (1), which also avoids the usual interpretation problems of the white-noise process. The same Γ as in the single-walker case is used, which is justified since the walkers are noninteracting. The additive, spatially uncorrelated, spontaneous emission noise $s(x, t)$ is a Poissonian process whereby ϕ is increased by ε at random times, whose separation τ is distributed as $\gamma \exp(-\gamma\tau)$. The process $v(x, t)$ is Gaussian distributed with

$$\langle v(x, t)v(x', t') \rangle = D\ell\Delta(x - x')\delta(t - t'), \quad (9)$$

where the spatial scale ℓ is introduced for dimensional consistency. We further assume that this noise has a finite

correlation in space, described by the smooth function $\Delta(x)$, an assumption often done in other contexts [27] to ensure that the corresponding term is well defined.

The mean-field equation obtained by neglecting fluctuations in Eq. (8) and replacing s by its average γ is a simplified, one-dimensional version of the Letokhov equation for random lasers [2]. The steady state solution $\bar{\phi}(x)$ is not identically zero due to the γ term. Neglecting this, we obtain that $\bar{\phi}$ destabilizes for $\gamma = \gamma_c$, thus defining a threshold as assumed above [28].

As usual for a stochastic partial differential equation [30], Eq. (8) is intended as a limit of some discretization on a finite mesh whose spacing we denote by Δx . For definiteness and for actual numerical investigation, we choose the discretized equation for ϕ_i to be [$i = 0, \dots, N + 1, (N + 1)\Delta x = L$]

$$\dot{\phi}_i = D[\phi_{i+1} + \phi_{i-1} - 2\phi_i] + \Gamma(\phi_i) + \frac{1}{2}(v_{i+1}\phi_{i+1} - v_{i-1}\phi_{i-1}) + s_i, \quad (10)$$

with v_i being independent Gaussian variables with $\langle v_i \rangle = 0$ and $\langle v_i^2 \rangle = D\ell$ (we set $\Delta x = 1$). Such discretization is legitimate since $\Delta(x)$ is assumed to be smooth over distances of the order of the mean free path. The boundary conditions are $\phi_0 = \phi_{N+1} = 0$, and we also impose $v_1 = v_N = 0$ to ensure that the multiplicative term conserves the total energy $\sum_i \phi_i$. For the time derivative, we use a simple Euler discretization with a time step Δt .

In the simulations, we choose s_i to be either a Poisson process (i.e., $s_i = 1$ or zero with probability $\gamma\Delta t$ and $1 - \gamma\Delta t$, respectively) or a Gaussian with the same average, $s_i = \gamma + \sqrt{\gamma}r_i$, r_i being normally distributed and independent random numbers with $\langle r_i \rangle = 0$ and $\langle r_i^2 \rangle = 1$. Although the first choice is closer to the original formulation of the model, we found that in practice the two processes yield almost indistinguishable results.

The term in v of Eq. (8) is the leading stochastic correction to the mean-field evolution. It is a multiplicative process and can be regarded as a kind of random advection whereby fluctuations are transported almost coherently while keeping the total energy conserved. It thus acts against the diffusive term that tends to smooth out fluctuations. As a result, the field is highly intermittent in time, with large-amplitude bursts emerging from a lower-amplitude background (Fig. 1). It is thus intuitively plausible that such a term is responsible of most of the nontrivial statistics of the field. As a matter of fact, random advection dynamics is known to yield strongly non-Gaussian distributions, and even power-law tails [31]. Actually, equations similar to (10) (with linear gain and periodic boundary conditions) have been studied in Refs. [32,33] to model an active scalar (e.g., a temperature field) convected by a random velocity field. It was argued that power-law tails generically arise when, as here, both multiplicative and additive noises are present. Actually, there are two sources

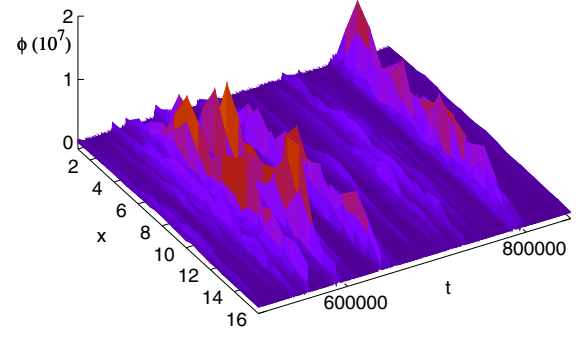


FIG. 1 (color online). Intermittent evolution of the intensity $\phi(x, t)$ obtained by numerical integration of Eq. (10): $L = 16$, $\gamma = 1.02\gamma_c$, $D = 1/2$, $\ell = 1$, $\Gamma(\phi) = \gamma\phi/(1 + \phi/\phi_s)$, $\phi_s = 10^8$. Here and in the following $\Delta t = 0.01$, s_i is Poissonian (see text).

of additive noise. One is the spontaneous emission term s ; the other stems from the fact that the deterministic, steady state value is $\bar{\phi} \neq 0$, thus yielding an additive contribution of order $\partial(v\bar{\phi})/\partial x$. There is, however, a crucial difference, dictated by the physical origin of the noise: the advective term is a finite-size effect which decreases with the system size. This can be demonstrated by dimensional analysis. To be more general, let us consider the extension of Eq. (8) to d dimensions,

$$\frac{\partial \phi}{\partial t} = D\nabla^2 \phi + \gamma\phi + \nabla \cdot (\mathbf{v}\phi) + \gamma, \quad (11)$$

where \mathbf{v} is a d -dimensional vector whose components v_ν ($\nu = 1, \dots, d$) are Gaussian distributed and satisfy a relation akin to Eq. (9),

$$\langle v_\nu(\mathbf{x}, t)v_{\nu'}(\mathbf{x}', t') \rangle = D\ell^d \delta_{\nu,\nu'} \Delta_d(\mathbf{x} - \mathbf{x}') \delta(t - t'), \quad (12)$$

with Δ_d being a generalization of the function in Eq. (9). For simplicity, we neglected again the $\sqrt{\Gamma}$ term, replaced s by its average γ , and restricted to the simpler case $\Gamma(\phi) = \gamma\phi$. Let us rewrite Eq. (8) introducing the dimensionless variables $\mathbf{x}/L \rightarrow \mathbf{x}$, $Dt/L^2 \rightarrow t$, $\phi/\varepsilon \rightarrow \phi$, and we also rescale $D\gamma/L^2 \rightarrow \gamma$. The noise rescales accordingly as $\sqrt{L^{d+2}/D\mathbf{v}} \rightarrow \mathbf{v}$. Thus, Eq. (8) is written in the dimensionless form

$$\frac{\partial \phi}{\partial t} = \nabla^2 \phi + \gamma\phi + \lambda(L)\nabla \cdot (\mathbf{u}\phi) + \gamma, \quad (13)$$

where each component of \mathbf{u} is again an uncorrelated Gaussian variable with $\langle u_\nu^2 \rangle = 1$ and we have made explicit the rescaled noise strength which turns out to be $\lambda \propto (\ell/L)^{d/2}$. As a consequence, we expect that the multiplicative noise will yield sizable power-law tails only for small enough sizes. When $L \rightarrow \infty$, the diffusive term in Eq. (11) dominates and the power law is no longer observable [33].

The numerical simulations of Eq. (10) are in qualitative agreement with the above argument. We monitored the distribution of the field at the center of the mesh $\phi_{N/2}$ as

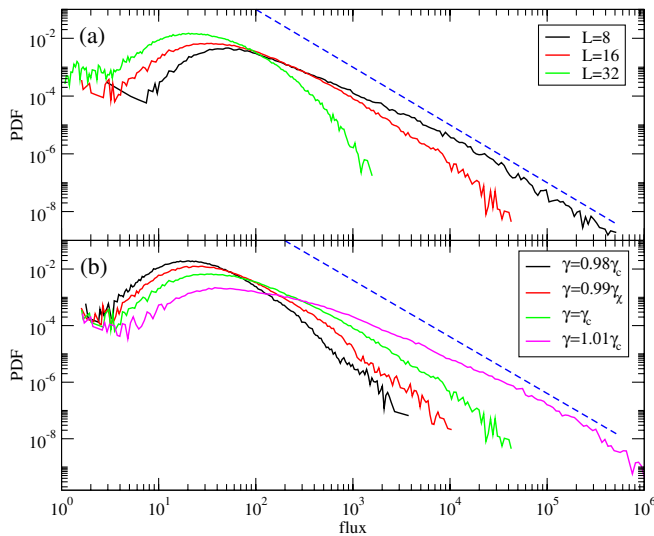


FIG. 2 (color online). Probability distribution functions (PDF) of the flux J for (a) $\gamma = \gamma_c$ and increasing L values (right to left) and (b) $L = 16$, increasing γ values (left to right). The straight dashed line corresponds to the power law $\alpha = 1$ expected from Eq. (6). Flux data have been binned over consecutive time windows of duration $T_W = 10$, over a run of about 10^6 time units. Additive noise is Gaussian (see text); other parameters as in previous figure.

well as the outgoing flux $J = D[\phi_1(t) + \phi_N(t)]/2$ as a function of time. This quantity is of experimental interest, being related to the emission spectra. The tail of the distributions of both observables displays, upon increasing L , a smooth crossover from a power-law tail with an exponent α very close to the one predicted by Eq. (6) to a faster decay [see Fig. 2(a)]. The same occurs for fixed L upon approaching the threshold γ_c [Fig. 2(b)].

The existence of fat tails is intimately related to the possibility for a spontaneous fluctuation to grow well beyond the average. The indicator to quantify this is the generalized Lyapunov exponent [34]. For a perturbation $\delta\phi(x, t)$ of the field, which evolves according to the linearized version of Eq. (8), let $R(\tau) = \|\delta\phi(t + \tau)\| / \|\delta\phi(t)\|$ be the response function after a time τ to a disturbance at time t . The generalized finite-time Lyapunov exponent $\Lambda(q)$ is then defined by $\overline{R^q(\tau)} \sim \exp[\Lambda(q)\tau]$, where the over line denotes a time average. If $\Lambda(q) > 0$ for large enough q , then the system has a finite probability that a small perturbation results in a large one. Moreover, the deviations of $\Lambda(q)$ from a linear behavior in q are a measure of intermittency [34]. Figure 3 shows that $\Lambda > 0$ for $q > q_*$, with q_* increasing with L . This signals that events far from the average become increasingly rare, in agreement with the above dimensional arguments. Also, for $L = 8$, $q_* = 0.92$, which, according to Ref. [32], would yield a power-law decay with an exponent $1 + q_* = 1.92$, in agreement with data in Fig. 2.

To summarize, we have presented a simple model for diffusive random media with gain which yields a

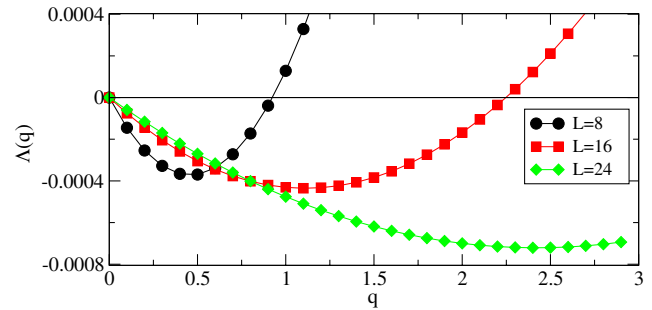


FIG. 3 (color online). The generalized finite-time Lyapunov exponent $\Lambda(q)$ for $\tau = 200$ and different system lengths L .

power-law distribution of the intensities. Our main result is the Langevin equation for the energy density field, Eq. (8), that establishes a novel connection between the physics of scattering media with gain (e.g., random lasers) and the theory of nonequilibrium phenomena in spatially extended systems. The random-advection, multiplicative term in Eq. (8) competes with diffusive and gain terms and is responsible for unconventional fluctuations. Its relevance is gauged by the effective noise strength $\lambda(L)$. This means that Lévy-like fluctuations are more likely to be observed in lower dimensions (for instance, in $d = 2$) and when the mean free path of photons is not too short with respect to the sample size. In other words, as in the case of advective mixing [32], small systems are also necessary to observe the effect. For an experimental validation of the model for random lasers, one should compare the emission statistics for samples with different $\lambda(L)$. This is achieved, for instance, by changing the gain volume (controlled by pumping) and/or the mean free path which can be tuned by varying the density of scatterers.

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