Lévy exponents as universal identifiers of threshold and criticality in random lasers

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Critical excitation in random lasers under picosecond and nanosecond pumping was experimentally studied. The resulting emission intensity statistics were analyzed using fits to α-stable distributions. We find that the transition value of α, the tail exponent of the stable distribution, is a clear indicator of the threshold of random lasing. We discuss this exponent as an identifier of the threshold. This definition is compared with the conventional definitions for the threshold, namely, the probability of random lasing in the case of coherent random lasers and the intensity enhancement and bandwidth collapse for diffusive random lasing emission. We find a universal applicability of the α exponent as an identifier of the threshold, and hence the criticality, in random lasers.

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I. INTRODUCTION

Random lasers are complex systems that realize amplification of light akin to conventional lasers, albeit depending on multiple scattering for the necessary feedback [1–5]. The generality of these conditions and the relative ease of achieving them have resulted in a vast body of experimental and theoretical research [6–23]. Among the various intriguing properties of random lasers, one that remains fundamentally relevant to all studies is the random lasing threshold [4,5]. Under conditions of population inversion, spontaneously emitted light undergoes coherent amplification when the multiply scattered light is trapped sufficiently long and travels a distance equal to or larger than the gain length. Critical excitation occurs when the realized inversion creates gain that equals the inherent losses in the system [4]. At the critical excitation, the system crosses the threshold beyond which the emission characteristics change.

In conventional lasers, the emission intensity diverges at threshold excitation, which is the universal way of identifying criticality [24]. In the case of random lasers, the indicators used for identifying the threshold are diverse, reflecting the variations in the configuration of random lasers. For instance, in powder lasers, transitions between electronic states offer gain which realizes scattering-aided stimulated emission [25]. Here, the intensity enhancement is used as the identifier. In organic gain media, broadband dyes are typically used for amplification. Here, both intensity enhancement and bandwidth collapse help in determining criticality [5]. In the random system, strong fluctuations in both intensity and bandwidth are a commonly observed behavior [26–28]. These fluctuations are largest close to the threshold, as shown from studies of the Pearson correlation between the pump energy and the peak emission intensity [29]. In such cases, a probabilistic measure, such as the probability of random lasing, has been employed to identify the threshold [30]. As an alternative measure, the ratio of the variance to the mean of the intensity has also been utilized [31]. Each of these measures is specific to the sample used or the excitation protocol employed in the system. The varying identification schemes pose a challenge in enabling comparative studies across differing samples or across various reports in the literature.

In that regard, we discuss a statistical measure that we observed in our recent experimental studies aimed at quantifying intensity fluctuations [32,33]. In recent years, significant interest in the Lévy fluctuations (power-law tailed) of random laser intensity has been generated [32–36]. Depending upon parameters such as pump energy, system size, disorder strength, etc., the fluctuations exhibit Lévy or Gaussian statistics. Since the emission involves mixed statistics, the method of histogram-fitting fails in the goal of regime identification. To that end, we imported an econophysical function for fitting the statistical data, namely, the α-stable distribution [37]. This function utilizes four parameters to completely describe a heavy-tailed distribution. The tail exponent α ∈ (0,2) describes the rate at which the tails taper off, with α = 2 indicating Gaussian behavior and α < 2 indicating Lévy behavior. The other three parameters are the location parameter μ, the skewness parameter β, and the width σ, which respectively describe the asymmetry, the mean, and the width of the distribution. Due to the lack of general closed-form expressions, the α-stable distribution is conveniently described by the characteristic function φ(t), i.e. the inverse Fourier transform of the probability density function, a particular parametrization given by [38]

\[ \ln \phi(t) = -\sigma^\alpha |t|^\alpha \left( 1 - i\beta \text{sgn}(t) \tan \frac{\pi \alpha}{2} \right) + i \mu t \quad \text{for } \alpha \neq 1, \]

\[ \ln \phi(t) = -\sigma |t|^2 \left( 1 - i\beta \text{sgn}(t) \frac{2}{\pi} \right) + i \mu t \quad \text{for } \alpha = 1. \]

To estimate α, we employed the method presented by Koutrouvelis [39], which is a regression-based technique that iteratively converges to a solution from a given initial estimate. For faster convergence, the initial estimates are calculated using the quantile-based McCulloch method [40]. Based on this numerical treatment of experimental data, we previously identified the Gaussian and Lévy statistical domains in random lasers [32] and thereafter also reproduced the first transition of the system from the Gaussian regime into the Lévy regime [33]. We also found that this transition corresponds exactly with the onset of random lasing. Thus, the α exponent itself can be an identifier of the lasing threshold and thereby the

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criticality in the system. In this Brief Report, we describe our experiments on dye-scatterer-based random lasers, studied under picosecond and nanosecond pumping. In each case, the related parameters are compared with the $\alpha$ exponent for identifying criticality. We show identical behavior between the $\alpha$ and the conventional identifiers over a span of the system parameters. Subsequently, we discuss the pros and cons of using this identifier for threshold.

II. PICOSECOND PUMPING

The sample under study was a suspension of ZnO nanoparticles ($d \sim 20$ nm) in a 2.5 mM solution of Rhodamine 6G in methanol. The density of scatterers was varied to span a range of disorder strengths characterized by the transport mean free path $\ell^*$ measured using the coherent backscattering technique. The sample was pumped with a pulsed Nd:YAG laser ($\lambda = 532.8$ nm, pulse width $\sim 30$ ps, pump spot diameter $\sim 70 \mu m$), and the emission was collected using a 50-cm spectrograph with an intensified charge-coupled device (ICCD) attached to it. The energy was varied sufficiently around the threshold pump energy $E_{th}$ to cover all the statistical regimes of emission. For each pump energy, a total of 2000 spectra were captured for efficient statistical analysis.

Figure 1(a) depicts typical spectra observed from samples with $\ell^* = 190$, 350, and 1500 $\mu m$ at varying pump energies. The lowermost blue curves represent the spectra at $E_p \sim 0.5 \mu J$, where the trivial fluorescence spectrum is observed, implying subthreshold emission. The upper red and black curves represent the spectra at stronger $E_p$, where larger spectral fluctuations are evident. The ultranarrow peaks diminish as the disorder strength increases. This is a consequence of the strong redistribution of gain among the larger number of open modes. This prevents a single or very few modes from having exceedingly large gain. Further, the output energy is larger for the weak-scattering sample, $\ell^* = 1500 \mu m$, compared to the one with $\ell^* = 460 \mu m$. This apparent contradiction can be understood from the strong angular dependence of emission in the backward direction in scattering systems. In weak disorder, the emission is restricted to a small cone in the backward direction, while it is more uniformly distributed in the stronger samples. Thus, the detector receives a larger signal from the weaker-scattering sample. For statistical analysis, we chose to operate at the wavelength $\lambda = 557$ nm. Two thousand intensity values at any given excitation energy are studied. The markers in Fig. 1(b) depict the intensity distribution for the three disorder strengths. Solid lines are the $\alpha$-stable fits for the data. The fits yielded the tail exponents $\alpha = 1.85, 1.45$, and 0.9 for $\ell^* = 190 \mu m$, $\ell^* = 350 \mu m$, and $\ell^* = 1500 \mu m$, respectively.

We choose a representative system with $\ell^* = 1500 \mu m$ to depict the behavior of the threshold identifiers across pump energies. A well-known statistic used in the case of coherent random lasers is the probability of random lasing $P_{RL}$ [30,41]. This defines the ratio of the number of spectra with ultranarrow modes to the total number of spectra captured. This definition originates from the observation that coherent modes are not excited at every pulse in the experiment; the gain has to be sufficient for the modes to be excited. The solid blue circles in Fig. 2(a) represent this probability, which becomes nonzero at $E_p \sim 0.6 \mu J$, identifying the threshold pump energy. Beyond this, a steady rise in $P_{RL}$ can be seen up to $E_p \sim 1.65 \mu J$, whereafter every spectrum possesses coherent modes [42]. Roughly, three domains can be identified in this behavior (depicted by magenta dotted lines), suggested by the monotonicity of $P_{RL}$. The first domain is where the probability is zero, the next is where it rises above zero to reach almost 1, and the third is where it remains close to 1. The solid red triangles in Fig. 2 represent the fluctuation coefficient $\Phi(I_\lambda)$, which is the ratio of the standard deviation to the mean of the intensities $I_\lambda$. $\Phi(I_\lambda)$ starts rising above 0.2 around $E_p \sim 0.55 \mu J$, suggesting an increase in fluctuations as the gain sets in. Observing the monotonicity of $\Phi(I_\lambda)$, its behavior can also be similarly demarcated into three domains; namely, $\Phi(I_\lambda)$ rises steadily, it hovers close to a maximum value, and it reduces steadily. In both parameters, the boundaries between the domains agree, clearly because the underlying physics remains the same. The solid black squares in Fig. 2 represent the Lévy exponent $\alpha$, which shows a transition from a Gaussian ($\alpha = 2$) to a Lévy behavior around $E_p \sim 0.6 \mu J$. This is, again, a signature of random lasing. Clearly, $\alpha$ is quite nonmonотonic. Hence, its progression does not identify with the behavioral domains marked earlier. However, it can be observed that fluctuations maximize and $\alpha$ minimizes at the same $E_p \sim 1.0 \mu J$. Importantly, the system first enters Lévy

![Figure 1](image-url)
FIG. 2. (Color online) (a) Statistical behavior of the random laser with \( \ell^* = 1500 \, \mu\text{m} \). Solid blue circles: \( P_{RL} \), probability of random lasing. Solid black squares: tail exponent \( \alpha \) of intensity distribution at \( \lambda = 557 \, \text{nm} \). Solid red triangles: fluctuation coefficient \( \Phi(I_\lambda) \). A coincidence of \( E_p(0.6 \, \mu\text{J}) \) at which \( P_{RL} \) becomes nonzero and \( \alpha \) falls below 2 can be observed. (b) Threshold pump energy \( E_{th} \) estimate using \( P_{RL} \) (blue circles) and the \( \alpha \) exponent (black squares).

regime at the same \( E_p \) where the second domain for the other two parameters begins.

\( \Phi(I_\lambda) \) suffers from the disadvantage that there is no absolute value that can be identified for the threshold. In contrast, \( P_{RL} \) rises above zero at the threshold, while \( \alpha \) drops below 2, both of which are definitive numerical conditions. Therefore, we compare the behavior of these statistics in the subsequent analysis. Figure 2(b) depicts this comparative study of estimates of the threshold pump energy \( E_{th} \). The probability of random lasing is not defined for \( \ell^* \) of 190 \( \mu\text{m} \) because of the absence of any coherent peaks in the emission spectra at any \( E_p \), which is evident from Fig. 1. Clearly, the Lévy exponent \( \alpha \) estimates the threshold \( E_p \) in accordance with the other statistic. Further, it can be applied at \( E_p \) where \( P_{RL} \) is not applicable.

### III. NANOSECOND PUMPING

Under long-pulsed pumping, the fluctuations in spectral line shape are minimal. However, the emission has intrinsic fluctuations in intensity upon crossing the lasing threshold. We performed experiments similar to the one above using an Nd:YAG laser with a pulse width of 5 ns. Typical emission spectra collected from the samples with \( \ell^* = 330 \) and 1500 \( \mu\text{m} \) under nanosecond pumping are depicted in Fig. 3(a). The lowermost blue curves represent the spectra before the system crossed the lasing threshold, and the upper red and black curves show spectra at an energy just above the lasing threshold. Here, the threshold was chosen using the conventional method of identifying \( E_p \) where the output intensity diverges. Very mild line-shape fluctuations were observed in only the weak-scattering systems. Distinct ultranarrow peaks were not observed in any condition, confirming that the emission was restricted to diffusive random

FIG. 3. (Color online) (a) Typical spectra from systems with \( \ell^* = 330 \) and 1500 \( \mu\text{m} \) under nanosecond pumping. Lowermost blue curves: subthreshold \( (E_p \sim 15 \, \mu\text{J}) \) spectra. Upper red and black curves: spectra at \( E_p \sim 70 \, \mu\text{J} \), above the random lasing threshold. (b) Parameters for a representative system with \( \ell^* = 800 \, \mu\text{m} \) as a function of \( E_p \). Blue circles: bandwidth collapse. Black squares: tail exponent \( \alpha \). Red triangles: \( \Phi(I_\lambda) \). Orange stars: emission intensity \( I_{out} \). (c) Comparison of threshold energies \( E_{th} \) estimated using the input-output curve and \( \alpha \) statistics.
lasing. As can be observed, the FWHM of the lasing spectra is much smaller.

We studied the increase in fluctuations and intensity enhancement over a range of \( E_p \) in a system with \( \ell^* = 800 \mu \text{m} \), as shown in Fig. 3(b). The orange stars depict the emission intensity \( I_{\text{out}} \) as a function of \( E_p \), showing a rapid increase above \( \sim 25 \mu \text{J} \), indicating the lasing threshold. The red triangles depict the \( E_p \) dependence of \( \Phi(I_\text{in}) \), which increases to a maximum value of 0.2 at \( E_p \sim 70 \mu \text{J} \), after which it decreases. The maximal \( \Phi(I_\text{in}) \) is smaller (\( \sim 0.2 \)) in comparison to that in coherent random lasers (\( \sim 0.75 \)). These weaker fluctuations imply a weaker heavy-tailed nature of the distribution, which is reflected in the \( \alpha \) values depicted by black squares. We can see that the maximal \( \Phi(I_\text{in}) \) corresponds to the minimum \( \alpha \). The blue circles represent the bandwidth of the ensemble-averaged spectra. The bandwidth is seen to collapse to a value of \( \sim 9 \text{ nm} \) at \( E_p \sim 200 \mu \text{J} \). The bandwidth collapse is a continuous process, unlike \( I_{\text{out}} \), which shows a clear change in slope. A comparison of the threshold energy estimates using the \( \alpha \) statistics and input-output curve is shown in Fig. 3(c). Clearly, the agreement between the two curves shows that \( \alpha \) provides an excellent identification of the critical excitation.

An additional advantage of using \( \alpha \) as an identifier is that it can be defined for individual wavelengths. Indicators like intensity enhancement and bandwidth narrowing are, by definition, macroscopic and are defined over the entire spectrum. In typical random lasers, the gain medium is broadband, and modal overlap leads to gain competition between wavelengths. Every frequency mode is capable of broadband, and modal overlap leads to gain competition by definition, macroscopic and are defined over the entire

\[ \Phi(I_\text{in}) \sim \mu \text{m} \] (black circles) and \( \ell^* = 1500 \mu \text{m} \) (red squares). For both systems, the critical excitation remains flat over a range of central wavelengths and rises at the sides of the spectrum. The blue end of the spectrum experiences strong absorption and hardly reaches criticality. The threshold exists over a much narrower range under nanosecond pumping since mode competition drains out any gain from the wings of the spectrum. Such features are inaccessible with bandwidth and output intensity as the identifiers.

\[ IV. \text{ DISCUSSION} \]

In summary, we have discussed the \( \alpha \) exponent as an indicator of criticality in random laser systems. Random lasers are inherently statistical systems, so any relevant parameter needs to take the fluctuations into account. This criterion is satisfied by the \( \alpha \) exponent. Such a statistically consistent identifier precludes the subjectivity associated with threshold identification, such as that in the case of the probability of random lasing. Here, one has to visually identify an ultranarrow mode, which, so far, does not have a rigorous definition. There are no set rules as to how tall the ultranarrow modes need to be to count as random lasing modes. The intensity enhancement that occurs at the threshold is universally recognized for identifying the threshold. In the case of coherent random lasers, the output intensity strongly fluctuates, and an average intensity has to be calculated. In diffusive random lasers, the collapse of the bandwidth is a continuous process, and no absolute values can be assigned to identify when the gain has overcome the losses. Various different amplifying media have inherently differing fluorescence bandwidths. In addition, the rate of the bandwidth collapse is variable from sample to sample and depends crucially on the concentration of the dye and the focal spot size of the excitation laser.

Threshold identification using \( \alpha \) is difficult under conditions where fluctuations are weak and \( \alpha \) remains very close to 2. This situation occurs under nanosecond pumping with particularly large excitation spot sizes, resulting in self-averaging of the gain. In all other situations, \( \alpha \) can be a robust identifier of the threshold. Clearly, \( \alpha \) is not a readily available parameter like the intensity or the bandwidth and needs to be calculated. However, there are easily available routines that can be used to compute this parameter, so this difficulty can be overcome. This appears to be an acceptable limitation considering the advantages such as statistical consistency, objectivity of definition, and universal applicability regardless of experimental protocols. We hope this idea triggers more discussion of universalizing the parameters in diverse random laser systems.

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[42] At high $E_p$, multiple coherent modes average out, making them indistinct, but $P_{RI}$ is taken as 1.