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Review on Levy Statistical Distributions and its Physical Applications

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Abstract

Levy distribution is one of important classification of statistical distribution. he Lévy distribution, named after Paul Lévy, is a continuous probability distribution for a non-negative random variable. The present work explained the Levy statistical distribution and its application like quantum dots and random laser.The characteristic of the QD and random laser has been explained in detail.

In this work, a rigorous review of published literature regarding blinking QD and random laser has been presented.

1. INTRODUCTION TO LEVYDISTRIBUTION

In recent years the research in physics is not restricted to critical phenomena only. Efforts are being made to make it inter-disciplinary to have better inside of nature. The Statistical Mechanics is increasingly becoming a generalized tool for the simulation and explanation of diverse physical problems. In reference to above aspects we cannot deal with large numbers of quantum particles using the law of mechanics. This gives rise to the concept of statistical mechanics. Using statistical mechanics many experimental works on heat, sound, current carrier and light diffusion are successfully explained [1-3]. Interestingly, all of them used normal Gaussian random walk as the tool to explain the physical problems. In this case, for a single straight line motion, the probability distribution of free path length x was found to follow a Gaussian distribution. The consequence of Gaussian (classical) statistics is that the length wise distribution for N random motion is also described by a Gaussian function, but with the variance N times larger than the individual path length distribution. These theories also have been successfully explained through many observations in related field of chemistry, biology and economics[4-9].

As more and more artificial and natural systems were studied, the experimental results became more and more diverse and simple normal Gaussian distribution theory could not explain all the experimental results [10]. The phenomena that could not be explained by normal Gaussian statistics were called anomalous. A French scientist Paul Levy generalized the Brownian motion by considering other distributions for which one straight line motion and ***N*** straight line motion share the same mathematical form. The generalized term for them is “Stable distribution”. At a given temperature, these are called stable because, their average behaviors do not change with time (more and more straight line motion included). Stable distributions cannot in general be expressed as a closed domain analytically besides some specific cases. These three special cases are Gaussian, Cauchy and Levy distribution [11]. The theory, which governs large number of physical phenomena and enthused a large number of physicists, is their probability at higher coordinates, i.e. tail distributions. The above three distributions are quite different in this range, with Levy distribution having the fattest tail. Compared to Gaussian distributions, Levy distributions fall off as slowly at long distances. This non-Gaussian, heavy-tailed statistics is becoming a commonly used tool in several physical applications today [12]. Power-law distributions often appear in other physical phenomena that exhibit very large fluctuations, for instance the spectral fluctuations in random lasers, motion of charged particle in plasma, locomotion of moving animals and even the evolution of the stock market instability. This statistics is also proved to be a root cause behind the rare physical phenomena which cause ergodicity breaking (i.e. the ensemble average is no longer equal to the time average). Therefore, a detailed analysis of such statistics keeping a close observation on the experimental data is of great recent interest.

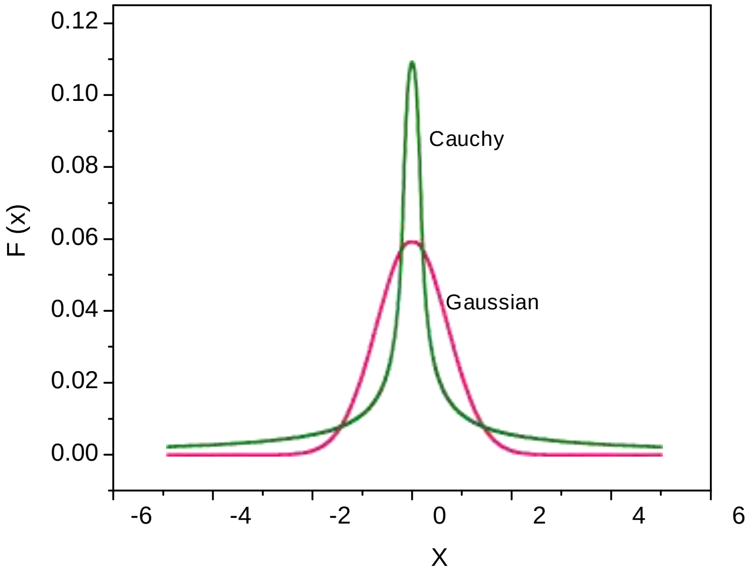

Fig. 1. A comparison of Gaussian and Cauchy distribution

2.0 APPLICATIONOFLEVYRANDOMWALK: BLINKING QUANTUM DOT

2.1 Introduction to Quantum Dot

In bulk semiconductors, electrons are free to roam about in randomly in three dimensions. However, interesting electrical and optical characteristics emerge when we confine the electrons to move in a lower dimension. Confining the electrons in two, one and zero dimensions give rise to Quantum Well (QW), Quantum Wire (QWire) and Quantum Dot (QD) respectively. This is achieved by sandwiching a lower band gap material in between higher band gap materials. Due to band gap differences, these structures cause the potential differences where by, the electrons are confined in the lower band gap materials. This confinement changes the density of state in the material and gives rise to interesting properties. The requirement is that, the size of the confined lower band gap material should be less than the De-Broglie wavelength of electrons in that material [13]. Figure 2 shows the schematic diagram for bulk, QW, QWire and QD. The density of states as a function of energy for them is also shown in the samefigure.

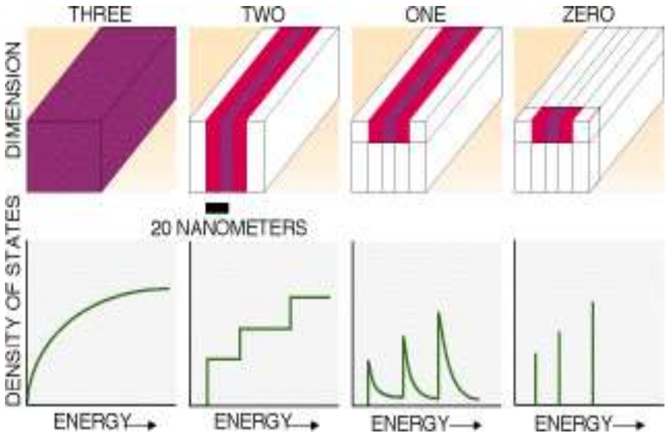

Fig. 2 Schematic diagram of (a) bulk, (b) quantum well, (c) wire and (d) dot materials with the density of states as a function of energy in those material.

2.2 Conceptualization of blinking properties of QuantumDots

When a photon of energy higher than the band gap energy of a semiconductor, is absorbed, it creates electron hole pair. Within the recombination times these photo-generated carriers recombine emitting light with certain efficiency (this is called fluorescence properties). The blinking of single atom was predicted by Niels Bohr based on his "energy quantization" theory. However, experimentally it was not observed until 1990, because it was not possible to trap a single atom. There is hardly any problem for explanation of such blinking fluorescence if we deal with a single atom or molecule. However, single molecule is able to absorb and consequently emits a photon in each time event. We may say that single molecule is a single absorber and single emitter (Raman Effect). Interruption of single molecule fluorescence happens when the molecule occupies non-radiative state, for instance the triplet state. However, single Quantum Dot comprising of thousands of atoms, is not a single absorber and more than one e–h pair can be created in such QD simultaneously. It is surprising to observe that even QD shows blinking fluorescence. That means when we record the fluorescence from a single QD, its intensity fluctuates. When the intensity falls below a certain threshold value, the QD are said to be in the "*off*" state and otherwise in "*on*" state. So, why should a QD blink? It will be explained in more detail in nextsection.

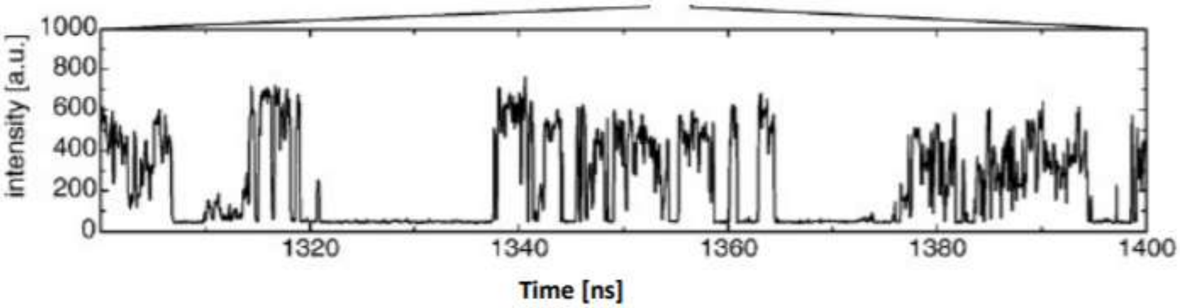

Fig.3. Intensity fluctuation of blinking QD.

Adding to the surprise, it was realized that the sojourn time viz. time spent in “*on”* and “*off”* states are not distributed exponentially which indicates some un-usual, non-poissonian kinetics [14, 15]. It was found that the PDF of “*on”* and “*off”* times follows power law behavior, with power law parameter varies between 1 and 2. The blinking characteristic of the QD has the following properties.

a. ***The existence of power laws from a threshold analysis:*** Blinking occurs over a wide range of timescales (µs to a few minutes). The shortest and longest times where set by experimental limitations. Distributions $P(t_{on})$ and $P(t_{off})$ can be fitted to powerlaws[16,17] of the form

$$1/t^{m_{on}} \text{ and } 1/t^{m_{off}}$$

Truncation times (“cutoffs”) were discovered in “*on*-time” distributions, beyond which the blinking statistics are not governed by power law [18]. Such cutoffs occur on the seconds timescale and may represent a competing physical process. This in turn interrupts power law blinking. A corresponding “*off*-t*ime”* cutoff in QDs has not been reported although it is speculated to occur on an hour timescale[19].

b. ***The existence of power law power spectral densities:*** Pelton and Guyot-Sionnest [20] have demonstrated that the power spectrum of the ensemble QD emission exhibits a power law form. This was also confirmed in the case of single QDs [20]. Furthermore, a kink in the power spectral density was recently observed with slopes reverting from ≈ *1* at low frequencies to ≈ *2* above 100 Hz[21].

c. ***The light-driven nature of the blinking process*****:** Intermittency is light induced, as indicatededby experiments revealing statistical "aging" of emission trajectories [22]. The ensembleemissionintensitydecaysundercontinuousexcitationandrecoversinthedark [19]. In existing experiments and theoretical understandings, the *on-time* distribution cut- off is inversely proportional to the excitation intensity [40, 41].

d. ***Exceptional lack of temperature dependence:*** *On/off* power law slopes are generally very weak temperature dependent between 10 K and 400 K [18, 25, 27]. This has led some investigators to speculate tunneling or another temperature independent physical processes at play. However, temperature sensitivity of on-time cutoffs has been observed by recently[12].

e. ***Relation between spectral diffusion and emission intensity :***Few authors have suggested that blinking is connected to another ubiquitous single molecule phenomenon: spectral diffusion [27]. Large shifts in the spectrum coincide with equally rare jumps of the intensity. This suggests a direct correlation between spectral diffusion and emission intermittency through the redistribution of charges on or nearby the QDsurface.

f. ***A continuous distribution of emission intensities and excitation lifetimes:*** Typical fluorescence intensity trajectories from single QDs do not mimic random telegramatic noise [28]. Thus unique "*on"* and "*off"* levels are not seen. Instead, many intensity levels exist. Schlegel and Mews [29] have since then found that such intensity fluctuations are correlated with changes in the QD emission lifetime. Additional few [20,21] have confirmed this and have, in turn, shown through simultaneous emission quantum yield measurements that non-radiative recovery rate fluctuations are connected to intensity variations. Furthermore, lifetimes much longer than the usual radiative lifetime of ~20 ns with a power-law distribution towards longer times have been observed [25]. Inliterature,

the results of the experiments are not well defined. A summary of the recent results reproduced in the following table clarifies this point

The following Table presents a review of publishedliterature in the field of blinking QD.

Table1. A review of published literature regarding blinking QD

Group (year)	Material	No. of atoms in QD	Radius (nm)	Temp. (K)	Intensity (kW/cm^2)	m_{on}	m_{off}
Verberk[24] (2002)	CdS	1	2.5	1.2	------	1.65	1.65
Brokmann[25] (2003)	CdSe-ZnS	215	------	300	------	1.58	1.48
Shimizu[9] (2001)	CdSe-ZnS	> 200	1.5	300,10	0.1-0.7	1.5	1.5
Kuno[16] (2005)	CdSe-ZnS	~200	1.7-2.9	300-394	0.24-2.4		1.5-1.75
Kuno[17] (2003)	CdSe-ZnS	> 300	1.7-2.7	300	0.1-100	1.9	1.54
Kuno[26] (2001)	------	~30	1.5	300	0.24	2	1.5
Chichos[27] (2004)	------	------	------	------	1.8-6.5	2.2	1.3
Hohng and Ha[28] (2004)	CdSe-ZnS	~1000	------	------	------	------	1.94-2.1
Muller[29] (2004)	CdSe-ZnS	41	4.4	300	0.025	1.55	1.05-1.25
Van-Sterk[30] (2002)	CdSe-ZnS	------	3.7	300	20	1.7-2.2	1.2-1.4
Kobitski[31] (2004)	CdSe	------	3.6	------	0.04-0.38	1.97-1.66	1.42-1.64

3.0 APPLICATIONOFLEVYRANDOMWALK: RANDOM LASER

3.1 Introduction to RandomLaser

A laser is usually constructed from two basic elements: a material that provides optical gain through stimulated emission and an optical cavity that partially traps the photons. When the total gain in the cavity is larger than the losses, the system reaches a threshold and lases. It is the cavity that determines the modes of a laser, that is, it determines the directionality of the output and its frequency. Random lasers work on the same principles, but the modes are determined by multiple scattering and not by a lasercavity.

Multiple scattering is a well-known phenomenon that occurs in nearly all optical materials that appear opaque. It is therefore quite common in daily life and determines the appearance of e.g., clouds, white paint, powders and even human tissue. Light rays that penetrate these materials are scattered often thousands of times in a random fashion before they exit again. This type of propagation is that of a random walk, just as in the Brownian motion of particles suspended in a liquid (Figure 2.4). The fundamental parameters describing this process are the *mean free path* (the average step size in the random walk) and the *diffusion constant*. Scattering in disordered optical material is complex yet completely coherent. This means that the phase of each of the optical wavelet undergoing a random walk is well defined and interference effects can occur, even if the material is strongly disordered. The most clear visualization of interference in multiple scattered light is laser speckle[1] [30], which exhibits grainy pattern when observed at a laser pointer that is scattered from, say, a piece of paper. The difference between light diffusion and multiple scattering is that diffusion refers to a simplified picture of multiple scattering in which interference effects are neglected.

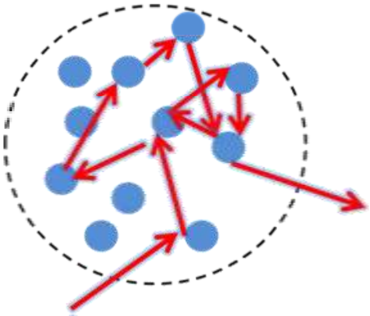

Fig.2.4 Multiple light scattering with gain.

A random collection of microspheres containing laser dye is excited (for example, by an external light source) to obtain population inversion. The microspheres then scatter light several times, as there is no outlet for ray. Incoming light is trapped and goes on making multiple reflections (scattering) till it is guided to leave cavity. Hence, it amplifies it in the process. The propagation of the light waves becomes that of an amplified random walk.

Multiple scattering due to randomness not only occurs in natural materials, but is also intrinsically present in photonic material, such as photonic crystals, intended as a component of optical devices. In those materials, multiple scattering has always been considered as an unwanted property arising from structural artifacts. It has now become clear that such artifacts are difficult to avoid and form a major industrial bottleneck [31]. Using multiple scattering to introduce new functionalities, therefore, opens up a completely new perspective on disorder in photonicmaterials.

3.2 Emission properties of a randomLaser

A better comprehension of a laser can be had by imagining it as an optical structure or artifact that satisfies the following two criteria :

(1) Light is multiple scattered, owing to randomness and amplified because of stimulated emission.

(2) There exists a threshold, due to the multiple scattering, above which total gain is larger than

totalloss.

This definition includes all multiple scattering systems with a gain in broad range of the mean free paths ***1*** , where there is no lower limit for ***1*,** but an upper limit exists which is approximately equal to the system size (otherwise the sample becomes transparent). Light diffusion with gain has already been discussed theoretically by Letokhov in the 1960s. who argued that for a diffusion process with amplification, occures in a situation where the total gain is proportional to the volume of the cavity and the losses are proportional to the total exposed surface [32]. It is then easy to see that there exists a critical volume above which gain becomes larger than loss, and the intensity reduced. If the gain depends on wavelength, this model would also predict that the emission spectrum narrows down above threshold with a maximum intensity at the wavelength of maximum gain. In addition, relaxation oscillations as well as laser spiking can be found in such a diffusive model [33].

Several of these features have indeed been observed in experiments. Briskina and co-workers [33] and later Migus and co-workers [34] could make a disordered amplifying material by grinding laser crystal into a fine powder. They managed to excite this powder optically and achieve optical gain through stimulated emission together with multiple scattering. Above a certain threshold gain level, the emission spectrum was observed to narrow down and the peak intensity to increase. A different strategy to achieve multiple scattering with gain was followed by Lawandy et al. [35], who suspended micro-particles in a laser dye. The advantages of such a material are that the amount of scattering can easily be varied by changing the particulate concentration and that the material is in a state of suspension (i.e. fluid from). Lawandy called this material laser paint, as it constitutes a laser that can be painted directly on a surface. Inearlywork, Lawandy did not always considered his explanation in the regime of random lasing. However, he also observed similar effects for single or very small particulate concentration [37, 38], which he corrected this in later work [40]. To call a material a random laser, the multiple-scattering process has to play a vital role in determining the lasing process.

There exists a misconception in the current literature that diffusive and coherent random lasers should be separately identified. However, such a distinction would suggest that in some material the light scattering process is subject to interference effects, whereas in others it is not. In all random-laser material, the multiple scattering processes are elastic, so that interference effects are present and remains an integral part of the physical problem. The question is whether the effect of this interference is observed in a specific experimental configuration. For instance, by using long excitation pulses or by averaging over several laser shots, some interference effects are averaged out. The remaining effects after averaging, such as a smooth narrowing of the spectrum, can be described by a simplified model of diffusion with gain that does not need to consider interference. It is however possible to observe from the same random-laser material either a smooth spectrum or narrow spikes. The latter can only be modeled by taking into account of interferenceeffects.

To model a *random laser* correctly requires solving Maxwell's equations for a system of randomly varying refractive index with a positive imaginary part. The optical modes that are found this way are complex and can be anderson- localized or otherwise confined in space or even extended, depending on the mean free path of the sample particulates. In addition, other properties can determine

1) The nature of the modes, such as the amount of (long-range) correlation in the refractiveindex.
2) The presence of partial order or stronganisotropy.

Given the broad scale of materials that is studied in the context of random lasing, it is desired to enrich, physics of amplifying random systems. With such diverse aspects it is pertinent to provide a clear definition of what is meant by a *randomlaser*.

3.3 Coherencecharacteristics of Randomlaser.

Coherent feedback is not required to obtain *random lasing*. The reason is that, like in a regular laser, it is not only the cavity itself that is essential for obtaining coherent laser emission, but also to understand this better. Hence it is desirable to know precisely first- and second-order coherence should be distinguished. First-order coherence is a measure of fluctuations of the field, whereas second order coherence accounts for fluctuations of the intensity. For a source of sufficiently narrow bandwidth, the first-order coherence is automatically high. Therefore, any mechanism that selects a specific narrow wavelength band (for example, a band-pass filter) creates first-order coherence [41]. Second-order coherence is more difficult to obtain owing to a tendency of photons to 'bunch', which creates large intensity fluctuations. In a laser, second-order coherence is obtained by saturation of the gain. This nonlinear effect limits the fluctuations of the intensity and thereby increases second-order coherence. In general if light is first order- and second-order coherent, the emission can conventionally be called 'coherent'. A laser cavity creates continuous (constant) reflections that there by form a convenient mechanism which automatically leads to the gain saturation required for second-order coherent light. However, thereareothersituationsinwhichgainsaturationcreateslightthathassecond-ordercoherence.

An example is that of the amplification in terms of number of photons by stimulated emission in system with population inversion. If the gain is large, the intensity will grow such that it depletes the gain medium completely. This suppresses the fluctuations of the intensity and there by gives rise to second-order coherence, or in other words 'poissonian'- photon – statistics that characterizes the coherent emission of a laser source. This also explains how a *random laser* can exhibit coherent emission, independent of the degree of localization of the modes and the amount of ‘coherent’ feedback.

4.0 CONCLUSION

The aim of this review is to present studies on levy distribution application like QD and random laser within the past years and see how is still changing. It is clear from the research reviewed that,

1. In the QD study; the PDF of “*on”* and “*off”* times follows power law behavior, with power law parameter varies between 1 and 2.
2. In the random laser study; it can exhibit coherent emission, independent of the degree of localization of the modes and the amount of ‘coherent’ feedback.

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