Chapter 1

An Introduction to Random Amplifying Media and Lévy statistics

This introductory chapter provides the background relevant to the experimental, theoretical and the numerical simulation work presented in the later chapters of the Thesis. The random amplifying medium (RAM) which has been a topic of intense research over the last decade is the object of our study. The first half of the chapter deals with RAMs, beginning with distinction of a RAM from a conventional laser, various length scales relevant to a RAM are defined, and regimes of scattering and localization discussed. Two types of RAMs are known in literature – the "dye-scatterer" RAM and the "crushed laser crystal" RAM. Our studies on both these RAMs are presented in the Thesis. The major emphasis is on the study of the statistics of fluctuations in the emission intensity from the different types of RAMs, over a wide range of parameters characterizing them. It is to be noted that, these fluctuations are over different microscopic realizations/complexions of the same macroscopic disordered (random) system. Normally, the fluctuations in physical systems follow the Gaussian statistics. However, we observe deviations from the Gaussian statistics in RAMs for a suitable choice of RAM parameters. The second half of the chapter introduces the elementary features of the Gaussian (section 1.3) and the Lévy statistics (section 1.4) to provide the required framework for discussing our findings. Section 1.5 discusses the distinguishing features of the Lévy and the Lognormal distribution. For completeness, some relevant statistical distributions are discussed in Appendix A (at the end of the Thesis).

1.1 Laser

The photon, being a massless boson, does not require conservation of number. This makes possible the amplification or absorption of photons. More specifically, a photon (unlike its fermionic counterpart, the electron), can stimulate an excited atom to emit another photon into the same electromagnetic mode (Fig. 1.1). Stimulated emission, predicted by Einstein in 1917, is the foundation for light amplification and oscillation (i.e., self-generation), and was first used by Townes [1] in the construction of a *Maser* (Microwave Amplification by Stimulated Emission of Radiation). The maser principle was later extended to the optical frequencies by Maiman [2], which led to the realization of *Laser* which stands for *Light Amplification by Stimulated Emission of Radiation*.

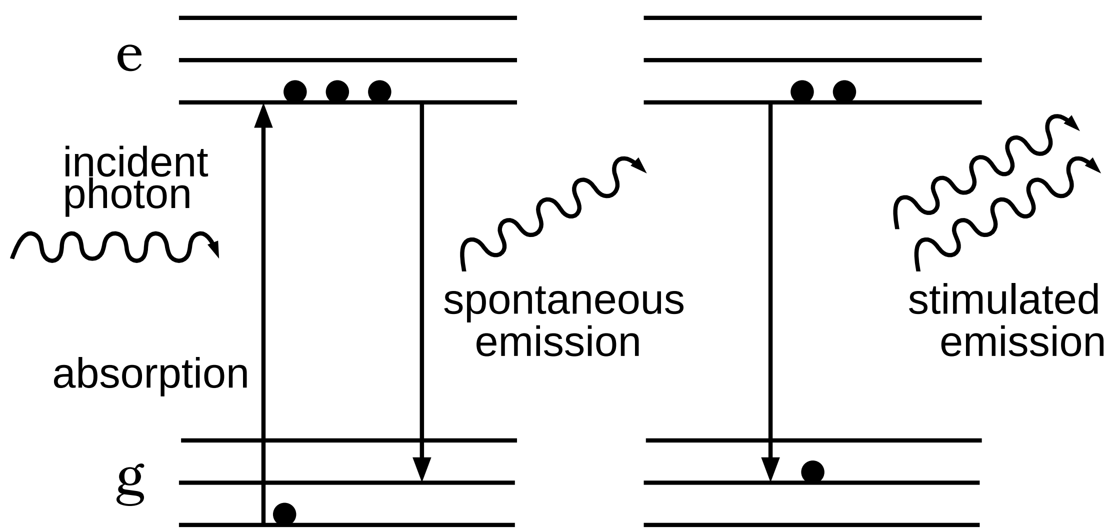

Figure 1.1: *Schematic of various absorption and emission processes: An incident photon causes an upward transition from the ground state (g) to an excited state (e), which results subsequently in the emission of a spontaneous photon. This photon then causes stimulated emission of a photon.*

A laser requires an optically active medium that amplifies light by stimulated emission, and a cavity, typically a pair of mirrors facing each other with the amplifying material enclosed in between, which provides a resonant coherent optical feedback for lasing. Coherent amplification over a specified band of frequencies is achieved by maintaining the active medium in a state of population inversion through a pumping mechanism. Threshold for lasing corresponds to a situation where the gain exactly balances the losses in the laser cavity. Thus, scattering is detrimental to laser action because it removes photons (the pump or the emitted) from the lasing modes of a conventional laser cavity, and destroys their spatial coherence, thus reducing gain.

As we shall see below, however, not only can one have lasing in the presence of scattering, the latter, can, in fact, enhance lasing.

1.2 Random Laser

Paradoxical as it may seem, when optical scattering is sufficiently strong in a disordered medium with gain, light scattering plays a positive role in both laser amplification and laser oscillation. In fact, in 1968, Letokhov [3] predicted theoretically that laser-like emission from amplifying disordered materials can be obtained using non-resonant positive feedback via multiple scattering of light. Such a disordered material with gain, which combines multiple scattering of light with amplification, is termed as “Random Amplifying Medium” (RAM). A RAM can be realized, for instance, by grinding a laser crystal into a fine powder and thus forming an aggregation of active scatterers which simultaneously amplify and scatter light, or by adding diffusely scattering passive particles such as polystyrene or rutile (TiO_2) microspheres to a laser-dye solution e.g., Rhodamine 6G dissolved in ethanol (Fig. 1.2). Instead of reflecting from one mirror to another as in a conventional laser cavity (Fig. 1.3(a)), in a RAM the light waves scatter randomly from one particle (scatterer) to another several times before they finally exit the RAM (Fig. 1.3(b)) [1]. Thus, in a RAM, scatterers

[1]The scattering off individual scatterers is not to be taken too literally. In the regime of a large number of sub-wavelength sized weakly scattering particles, and in the wave description of light, a scattering event is to be viewed as a change in the direction of propagation brought about by the collection of microspheres. Thus, the scattering length may exceed the mean distance between scatterers, and the location of a scattering event

play the role of mirrors in a conventional laser in providing an increased path length (or dwell time) to both the pump and the stimulated-emitted photons within the active medium – “mirrorless laser”. For example, in a dye-scatterer RAM, while the multiple scattering of pump photons results in their efficient absorption (Fig. 1.4) and hence, larger attenuation by the active (bulk) medium, the multiple scattering of emitted (fluorescent) photons (Fig. 1.2) in the active medium leads to their enhanced amplification by stimulation. In a RAM, the gain is given by $e^{\alpha l}$, where, α is the gain coefficient and l is the total path length traversed by the emitted photon in the active medium. The scattering is in random directions, and hence the name “random laser” for a RAM.

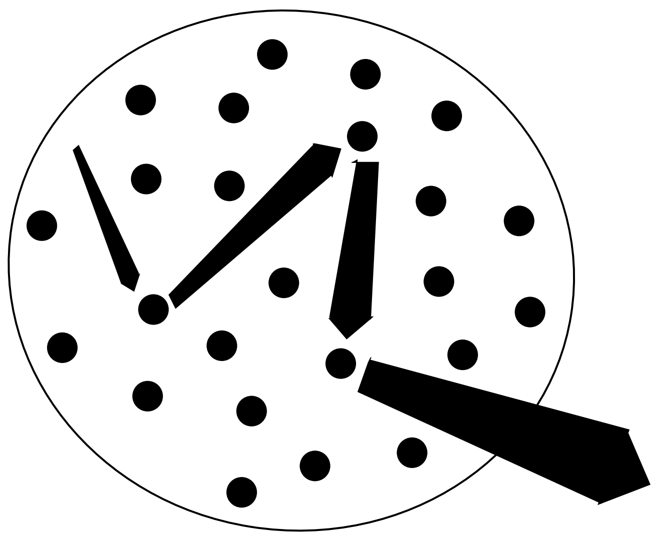

Figure 1.2: *Schematic of multiple scattering and amplification of a spontaneously emitted photon during its transit in the active bulk in a dye-scatterer RAM. (Line thickness is indicative of intensity).*

In a RAM, as in a conventional laser, the gain due to amplification competes with the losses. While the overall gain in a RAM depends on the excitation energy and on the scattering strength, the losses are due to absorption and the escape of photons from the amplifying medium. The threshold at which lasing occurs, indicated by the sudden drastic spectral nar-

need not literally coincide with the physical location of any single particle.

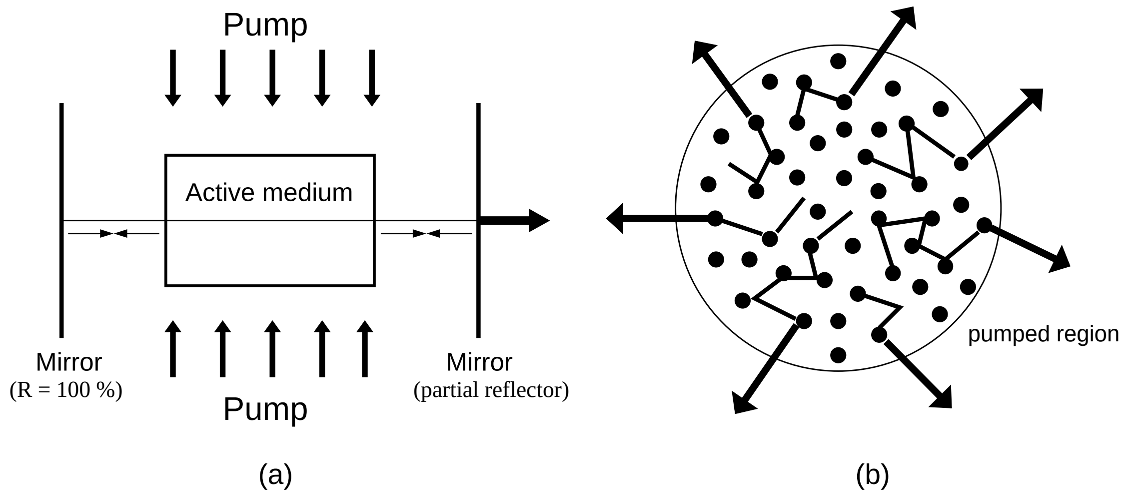

Figure 1.3: *Schematic of (a) Conventional laser, (b) Random laser*

rowing of the emission spectrum [4, 5] is determined by the balance between the gain and the loss, as in the case of a conventional laser [2].

Characteristic length scales for a random laser

In a RAM, light is both multiply scattered and amplified. The length scales relevant to a RAM are :

Scattering mean free path l_s :

l_s is defined as the average distance between two successive scattering events and is given by,

$$l_s = \frac{1}{n_s \sigma_s} , \tag{1.1}$$

where, n_s is the number density of scatterers and σ_s is the scattering cross-section of the individual scatterer. The scattering cross-section (σ_s) is given as, $\sigma_s = Q_s\, G = Q_s\, \pi\, (d/2)^2$: Q_s is the scattering efficiency and $G = \pi\, (d/2)^2$ is the geometrical cross-sectional area

[2]There is reason to believe that true lasing occurs when instead of a smooth peak with a width of a few nanometers, several extremely narrow emission peaks (width ~ 0.2 nm) appear in the spectrum. This is discussed, in detail, in chapter 2.

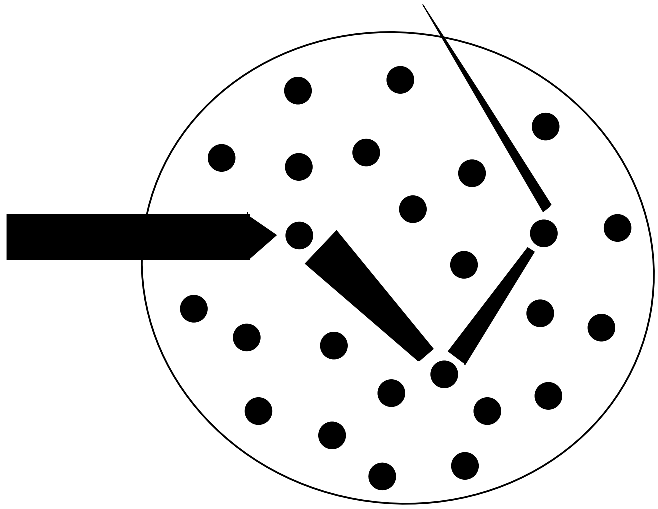

Figure 1.4: *Schematic of multiple scattering and attenuation of pump photons in a dye-scatterer RAM. These photons are absorbed by the dye molecules in the ground state, resulting in their subsequent excitation to the higher energy levels. (Line thickness is indicative of intensity).*

of the particle, d being the diameter of the scatterer. Thus, σ_s depends on the size of the scatterer, incident wavelength, and the refractive index mismatch between the scatterers and its surroundings.

Transport mean free path l_t :

The scattering mean free path, l_s, being inadequate to account for the anisotropy in scattering, a 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,

$$l_t = \frac{l_s}{1-g} , \tag{1.2}$$

where, $g =< \cos\theta >$ is the anisotropy parameter defined as the average cosine of the scattering angle θ. For isotropic scattering (e.g., Rayleigh scattering from particles of size $\ll \lambda$), $g = 0$ or $l_t = l_s$, while for complete forward scattering, $g = 1$.

Gain length l_g :

The gain length is defined as the e-folding path length in the active medium over which light intensity is amplified by a factor e.

Amplification length l_{amp} :

In a RAM, owing to multiple scattering and the consequent random paths of the photons, the actual arc-length of travel of a photon is much greater than the chord distance between the beginning and the ending points. This gives rise to the concept of l_{amp}, defined as the chord distance between the begin and the end points for which the arc path length traversed by the photon is l_g. In a homogeneous amplifying medium (without scattering), light travels in a straight line, thus $l_{amp} = l_g$. In the diffusive sample, $l_{amp} = \sqrt{D\tau_{amp}}$, where, D is the diffusion coefficient, $\tau_{amp} = l_g/v$, v is the transport velocity of light in the medium. In a three-dimensional system, $D = v\ l_t/3$, thus, $l_{amp} = \sqrt{l_t\ l_g/3}$. In an absorbing medium, the analogue of the gain length is the inelastic mean free path l_i, defined as the path length over which light intensity is reduced to $1/e$ of its initial value, due to absorption. Hence, the amplification length l_{amp} is analogous to the absorption mean free path $l_{abs} = \sqrt{l_t\ l_i/3}$.

Critical Volume/ Thickness :

For a RAM, critical volume is defined as the volume above which the system becomes unstable. For samples with slab geometry, critical thickness (instead of a critical volume) is defined as the thickness above which the intensity diverges and is given by
$L_{cr} = \pi\ l_{amp} = \pi\ \sqrt{l_t\ l_g/3}$.

Classifi cation of Random Lasers

As was discussed in the previous section, a random laser represents the process of light amplification by stimulated emission with the optical feedback provided by multiple scattering of light[6, 7]. Depending on the type of feedback, random lasers can be classified into two distinct categories : (i) random lasers with incoherent and non-resonant feedback, (ii) random lasers with coherent and resonant feedback. While, for the incoherent random lasers intensity feedback occurs which is phase insensitive (incoherent) and frequency independent (non-resonant), in the case of coherent random lasers, the field or amplitude feedback is operative, which is phase-sensitive (coherent), and therefore frequency dependent (resonant) – hence, the nomenclature.

We, first, discuss the "incoherent random lasers".

Random Laser with non-resonant (incoherent) feedback

In 1966, Ambartsumyan *et al.* realized a laser cavity in which one mirror of the Fabry-Perot cavity was replaced by a scattering surface which results in the multiple scattering of light in the cavity [8]. The feedback in such a laser serves merely to return part of the energy or photons to the gain medium – hence, energy or intensity (non-resonant) feedback. Due to the absence of resonant feedback, the cavity spectrum is not made up of discrete components at selected resonant frequencies; instead, it is a continuous one. With an increase of pumping intensity, the emission spectrum narrows continuously towards the center of the amplification line of the active medium. However, the process of spectral narrowing is much slower than in ordinary lasers [9]. Further, the emission of such a laser has no spatial coherence and is not stable in phase [10]. Therefore, this laser with a scattering reflector did not generate much interest in the scientific community.

In 1968, Letokhov [3] theoretically proposed self-generation of light in a RAM, in the diffusive regime (i.e. $\lambda \ll l_t \ll L$). By solving the diffusion equation for the photon energy density in the presence of uniform and linear gain, he showed that as the volume of the scattering medium exceeds the critical volume V_{cr} ($\approx (l_t l_g/3)^{3/2}$), the photon energy density increases exponentially with time. Because this process of photon generation is analogous to the multiplication of neutrons in an atomic bomb [11], this device is sometimes called a "photonic bomb". In reality the light intensity will not diverge (there is no explosion) because gain depletion quickly sets in and l_g increases.

For a long time Letokhov's theoretical work was not followed by experiments. In 1986, however, Markushev *et al.* showed that a powder of $Na_5La_{1-x}Nd_x(MoO_4)_4$ solid state luminophosphors under resonant pumping with short laser pulses (30 ns duration) at low temperature (77 K), could produce laser-like emission [12]. This was followed by extensive experimental and theoretical studies of stimulated emission in pulverized and highly scattering solid-state luminophosphors [13]-[19]. In a powder laser, it is hard to tell whether the feedback is provided by multiple scattering or internal reflection, because the gain medium and scattering elements are not separated in the powder. In 1994, Lawandy *et al.* [4] demonstrated isotropic laser-like emission from an optically pumped laser dye solution (Rhodamine 640 perchlorate in methanol), in which point-like scatterers (TiO_2 microspheres of mean diameter < 1 μm) were randomly suspended. A collapse of the linewidth of emission was observed above a threshold pump power, and was interpreted as the onset of lasing when the gain due to the enhanced path lengths within the active medium, brought about by multiple scattering, exceeded the intensity loss from the system. Line width collapse is a well-known phenomenon which occurs even in homogeneous amplifying media due to amplified spontaneous emission (ASE). But the remarkable aspect in these experiments was that the threshold of the pump power at which the emission spectrum collapsed drastically was almost two orders of magnitude smaller in the case of microsphere-laser dye suspension, compared to the case of ASE in the neat dye solution. Further, the peak emission intensity was shown to increase by more than three orders of magnitude upon addition of scatterers [5]. As opposed to the crushed laser crystal RAM, the dye-scatterer RAM separated the scattering and amplifying media thereby allowing the scattering strength to be varied independent of the optical gain via scatterer density and dye concentration respectively. This in turn facilitated a systematic study of the scattering effect on feedback. Since this observation, many experiments were carried out to examine the origin and various features of the narrow-linewidth emission reported by them. Simultaneously, several theoretical studies [20]-[25] have been conducted to provide models that will efficiently describe the behaviour of these materials and help in an understanding of the underlying mechanisms that are responsible for their laser-like characteristics.

Clearly, in all the above cases the phase condition within the scattering medium is ignored because of the non-resonant diffusive nature of feedback provided by the weak scatterers -

it only returns light into the gain volume, instead of to its original position. In fact, the probability of emitted light returning to its original position is so low in the diffusive regime ($l_t \gg \lambda$) that the interference effect on the feedback is negligible. Therefore, this kind of laser is called a random laser with non-resonant or incoherent feedback. Incoherent random lasing is similar to amplified spontaneous emission (ASE) in that it drastically narrows the emission spectrum (from a few tens of nanometers to a few nanometers) and transforms the usual linear excitation-emission intensity relation to a highly nonlinear one, above the pump threshold.

Now, we discuss the “coherent random lasers”.

Random laser with resonant (coherent) feedback

In the case of strong scattering and high optical gain coherent feedback can be obtained due to recurrent scattering over closed loop paths, (Fig. 1.5) where light returns to a scatterer from which it was scattered before. This is most probable for large scattering strengths (strong scattering), when the transport mean free path becomes comparable to the emission wavelength - $l_t \sim \lambda$ - the incipient photon localization regime. If the amplification along such a loop path exceeds the loss (due to escape from the loop), laser oscillation should occur in the loop, which then serves as a laser resonator. The requirement of the phase shift along the loop being a multiple of 2π (condition for constructive interference) determines the oscillation frequencies. Random lasing with coherent feedback is characterized by appearance of discrete, narrow lasing peaks (of linewidths < 1 nm, being limited by the spectrometer resolution) in the emission spectrum above the pump threshold in addition to a drastic increase of emission intensity.

Cao *et al.* demonstrated random lasing with coherent feedback in two distinct systems namely : (a) an aggregation of active scatterers, namely Zinc Oxide (ZnO) nanorod array and ZnO/GaN (Gallium Nitride) nanoparticles (semiconductor nanostructures) [26], and, (b) passive scatterers (ZnO particles) suspended in an amplifying bulk (Rhodamine 640 perchlorate dye dissolved in methanol) [27]. As remarked earlier, in the dye-scatterer system (b), the scattering and the amplifying media are separated which allows independent variation of

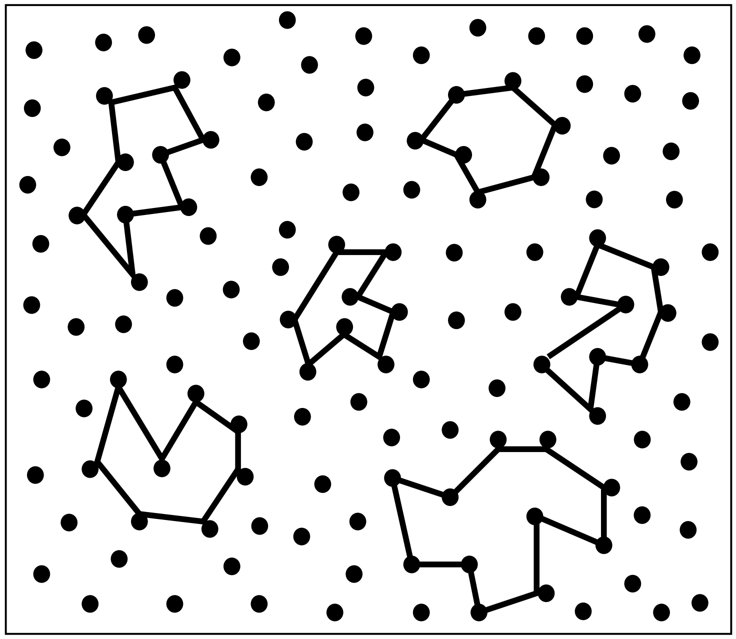

Figure 1.5: *Schematic of formation of closed loop paths in the random lasers with coherent feedback. These could well be a signature of the Ruelle-Pollicott resonance, as observed in the scattering of a particle moving in a medium of randomly positioned disks (in a plane) - the open n-disk billiard.*

scattering strength (via scatterer density) and optical gain (via dye concentration). However, in (a), the active scatterers act as both light scatterers and amplifiers. Further, these provide higher scattering strength owing to a larger contrast of refractive index and a higher density of scatterers. In both the cases, at low pumping power the spectrum consists of a single broad spontaneous emission band (linewidth $\sim$ a few tens of nanometers). When the pump power exceeded a threshold, discrete, narrow peaks (widths < 0.3 nm) emerged in the emission spectra in systems that were close to localization threshold (to be discussed shortly). The number of these discrete, spectral peaks increased with further increase in the pump power. Recurrent scattering and interference was invoked to explain the observed effects in these systems. There have, however, been reports of spiked emission from dye-scatterer RAMs, in the diffusive regime (well removed from the localization condition) [28].

The quantum statistical property of laser emission from the ZnO powder was also probed in a photon counting experiment [29]. It was observed that the photon number distribution in a single mode changes continuously from the Bose-Einstein distribution near the threshold to the Poisson distribution well above the threshold. Later, three dimensional, spatial confinement of laser light in a micron-size ZnO cluster (called a "micro random laser") was demonstrated, through multiple scattering and interference [30, 31]. The interference effect being wavelength sensitive, light at only certain frequencies could be confined in a given cluster.

While Cao *et al.* interpret their experimental findings as arising due to the formation of random laser cavities (virtual) due to recurrent scattering and interference effects in random media with discrete scatterers and strong short-range disorder ($l_t \sim \lambda$), referred to as "Quantum random lasers with coherent feedback", Vardeny and co-workers first demonstrated random lasing with coherent feedback in a different type of resonant cavity formed by smooth long-range inhomogeneity in a weakly disordered medium ($l_t \gg \lambda$), referred to as "Classical random lasers with coherent feedback". They conducted extensive experimental studies on random lasing in weakly disordered media such as π-conjugated polymer films [32, 33], organic dye-doped gel films [34], synthetic opals infiltrated with π-conjugated polymers and dyes [34, 35, 36], and found that at high excitation intensities a featureless amplified spontaneous emission band transforms into a finely structured spectrum having features as narrow as 0.1 nm. It was suggested that the long-range fluctuations of refractive index in their polymer films are most likely caused by inhomogeneity of the film thickness. Light is trapped in a high index region (which is much larger than l_t and λ), by total internal reflection at the boundary of this region.

In the classical random lasers with coherent feedback, formation of (closed) periodic orbits with small leakage results in light confinement. The interference effect plays a secondary role as it only determines the resonant frequencies in the periodic orbits. However, in the quantum type of random lasers with coherent feedback, the random media have discrete scatterers and strong short-range disorder, and thus the interference (incipient Anderson Localization) of scattered waves is essential to light trapping in a random medium.

Regimes of light transport in a random laser

Three regimes of light transport in a RAM can be distinguished depending on the strength of scattering :

(1) Ballistic regime

In this weak scattering regime, the size of the system L is comparable to (or smaller than) the transport mean free path i.e., $L \leq l_t$. Here, the only role of the particles is to scramble the directionality of the amplified spontaneous emission which would build up even in the absence of scatterers.

(2) Diffusive regime

In this case, $\lambda \ll l_t \ll L$, where, λ is the wavelength. While the first inequality ensures that localization effects are small, the second inequality implies multiple scattering of the wave traversing the system. In the diffusive regime, the strong, multiple scattering returns the photons to the active region thereby increasing their path length or dwell time in the gain region leading to enhanced amplification, compared to that in a clear sample without scatterers. This enhanced light amplification offsets the detrimental effects of diffuse, multiple scattering and other losses. The gain narrowing process will therefore be much more efficient; the presence of scatterers aids to narrow the spectrum of the output.

The propagation of light in the diffusive regime can be described as a random walk of photons with the direction of each succeeding step determined probabilistically, and can be described by the particle diffusion equation in a gain medium:

$$\frac{\partial I(\mathbf{r},t)}{\partial t} = D\nabla^2 I(\mathbf{r},t) + \frac{v}{l_g} I(\mathbf{r},t) \, , \tag{1.3}$$

where, $I(\mathbf{r},t)$ is the optical intensity at a position $\mathbf{r}$ inside the medium at time t, $D = vl_t/3$ is the diffusion coefficient, v is the transport velocity of light in the medium. The last term in Eq. (1.3) is the gain term. The diffusion equation is a classical equation that completely neglects interference effects between multiply scattered waves and describes only the average intensity. It may be noted in passing that a quenched disorder (of v/l_g) on a scale $\gg l_t$

can lead to the novel phenomenon of of intermittency - intense rare emission spots in the medium.

(3) Localization regime

For the case of wave propagation in strongly scattering media, interference effects can not be neglected. For example, interference between the counterpropagating waves in the highly disordered structure [37] enhances the intensity in the backscattering direction leading to the phenomenon of "coherent backscattering (CBS)" arising due to "weak localization" (Figs 1.6 and 1.7). The phenomenon of weak localization has been extensively studied in literature, both for the passive and the active random media [38]-[52].

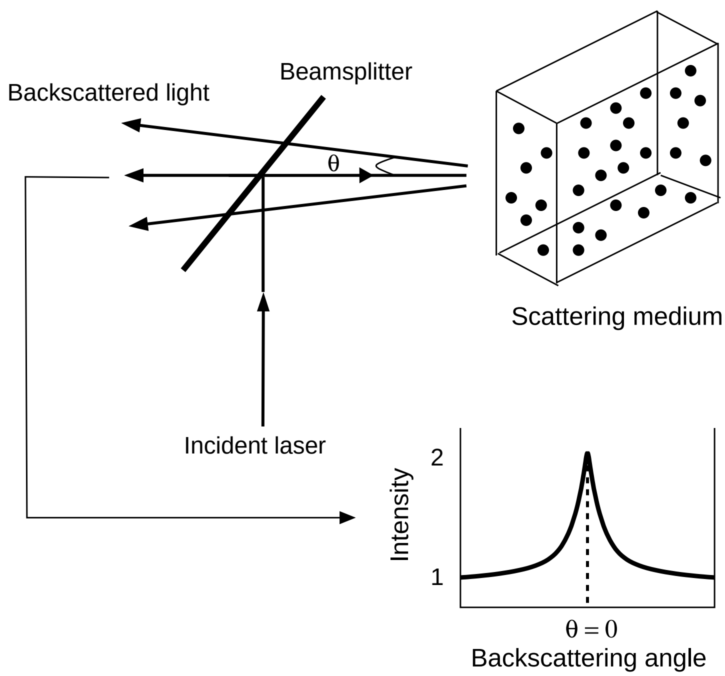

Figure 1.6: *Schematic of the experimental set-up used to record coherent backscattering. A beamsplitter is used to reflect the incident laser light onto the sample. The scattered light, transmitted through the beamsplitter is recorded, which gives the backscattering cone.*

When the scattering strength is increased beyond a critical value so that $l_t \leq 1/k$ (where, $k = 2\pi/\lambda$ is the wave vector) or $kl_t \leq 1$ – the Ioffe-Regel criterion [53, 54] – the system makes a transition into a localized state. This is referred to as Anderson (strong) localization

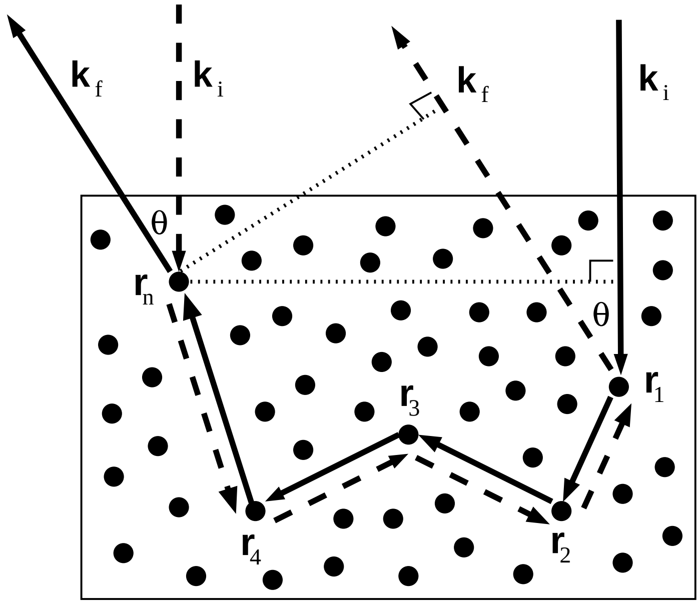

Figure 1.7: *Schematic of a scattering path (solid line) and its time-reversed mate (dashed line) with incident light direction* $\mathbf{k_i}$ *and final direction* $\mathbf{k_f}$. *Five scattering centers with positions* $\mathbf{r_1}$ *through* $\mathbf{r_n}$, *relative to an arbitrary origin. The scattering angle* θ *is defined relative to the backscattered direction* $-\mathbf{k_i}$. *The physical path length difference between the two time reversed scattering paths gives the optical path length difference.*

of light. Physically, this criterion states that localization occurs if the transport mean free path becomes comparable to the effective wavelength, so that the electric field can not even perform one oscillation before the wave is scattered again. Due to such strong scattering, the return probability of the intensity to closed loop paths is very high thereby reducing the diffusion constant. The diffusion constant can, thus, become zero at large scatterer strength, implying that the wave can no longer escape from its original region in space or the light propagation ceases and hence the name localization. Clearly, in the localization regime, light propagation is inhibited due to the interference of the multiply scattered waves. While the weak localization effect is already present in lower orders of multiple scattering, strong localization needs larger orders of scattering for which $kl_t \sim 1$. Thus, weak localization is said to be the precursor of the strong (Anderson) localization. Localization of light waves in strongly scattering media was theoretically predicted [55, 56, 57] in the 1980s which was then demonstrated experimentally (both in the microwave [58, 59] and the visible regime [60]).

Localization can also be understood in the mode picture. Theoretically, a localized state is an eigenstate of an infinitely large random medium. Due to the finite size of the medium and its open boundary, light leakage from the edges of the medium results in a finite lifetime of the localized state. This in turn leads to a spectral width of a mode, $\delta\nu$, which is the inverse of the escape time of photons from the medium/transit time through the sample (Thouless time). The Thouless number δ (or the inverse of the finesse factor for the virtual cavity, in the optics parlance) is defined as the ratio of spectral width of a mode $\delta\nu$, to the typical spacing between the modes $\triangle\nu$: $\delta = \delta\nu/\triangle\nu$. In the non-localization regime, quasimodes (eigen modes of the Maxwell equations in a passive random medium) overlap in frequency resulting in a continuous emission spectrum i.e., $\delta\nu > \triangle\nu$ so that $\delta > 1$. On the other hand, in the localization regime, quasimodes do not overlap, resulting in distinct, sharp, narrow spectral peaks such that $\delta < 1$. The localization threshold is set at $\delta = 1$ – the Thouless criterion [61]. Thus, the localization transition corresponds to a transition from overlapping modes to non-overlapping modes. While typical chaotic cavity lasers and photon localization lasers have non-overlapping modes ($\delta < 1$), for the diffusive random lasers quasimodes overlap ($\delta > 1$).

Light localization and coherent amplification

The coherent amplification (or absorption) of light has no counterpart in the electronic system as the latter, being fermionic, requires number conservation, while the former being bosonic, does not. Optical absorption hinders photon localization as it suppresses the interference effect of scattered light. Optical (coherent) amplification, on the other hand, enhances the interference phenomena in random media, thus facilitating light localization. However, the criterion for Anderson localization of light in an amplifying random medium remains to be developed. The very concept of describing the Anderson localization transition in terms of a vanishing diffusion coefficient as an order parameter, becomes questionable in active or dissipative media, where diffusion is not the only channel for change of the energy density [62].

Recently Chabanov *et al.* [63] developed a new criterion for photon localization in a passive and dissipative random media. They demonstrated that the variance of transmission fluctuation accurately reflects the extent of localization even in the presence of absorption. Unfortunately, this criterion does not seem to hold for active random media, because the variance of transmission fluctuation would diverge at the lasing threshold. Though, gain saturation prevents this divergence, the actual value of transmission variance depends on the saturation intensity which is determined by the material properties instead of wave transport. There is also doubt in applying the Thouless criterion for light localization in a passive random system to an active system. Though, there exists an ambiguity in determining light localization in amplifying random media, the effects of coherent amplification on light transport in disordered media has been extensively studied and compared with the effect of absorption [64]-[73].

Having discussed in detail about RAMs, their characteristic features and the present status of research on RAMs, the rest of the chapter will be an introduction to the relevant statistical background - the Gaussian and the Lévy statistics. We, first, introduce the Gaussian statistics.

1.3 Gaussian statistics

Random walk

A random walk, defined as a path composed of many independent random steps [74], is observed in a broad spectrum of systems ranging from watching winnings fluctuate in games of chance, to completely erratic motion of a drunkard wandering from one lamp-post to another, to modern studies of nonlinear dynamics.

Brownian motion, which describes the completely erratic motion of very small particles undergoing unending collisions with the surrounding particles, which hit them from all sides, is the simplest of random walks (Fig. 1.8). Fundamental dynamical processes such as molecular transport in liquids, atomic and molecular diffusion on surfaces, motion of microorganisms as well as many other examples have all been described in terms of Brownian

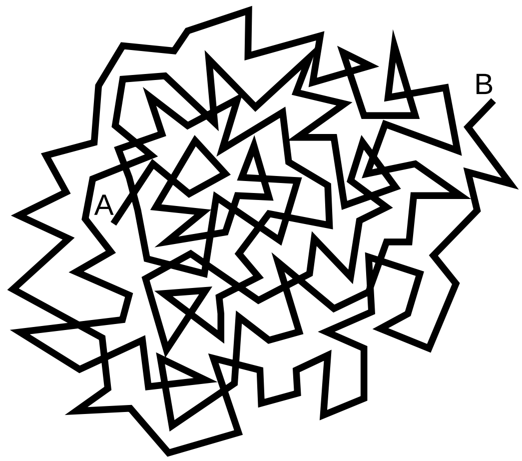

Figure 1.8: *Schematic of random walk (A $\rightarrow$ B) of a Brownian particle*

motion. Einstein showed that the mean-squared-displacement of the random walk of the Brownian particle grows linearly with time, i.e., $< x^2(t) > \sim t$; i.e., the root-mean-squared displacement of the Brownian particle from its starting point increases with the square root of time ($\sqrt{t}$), and not linearly. The probability of a Brownian particle being at distance d from its starting point after time t is represented by a “Gaussian”- a bell shaped curve centered at the origin (Fig. 1.9 : dotted curve) which very rapidly drops to zero. The width is proportional to $\sqrt{t}$, and is a measure of the distance beyond which there is little probability of finding the particle, i.e., as the particle goes far from the mean value, the probability rapidly drops to zero. A striking aspect of this result is its universality. Irrespective of the microscopic details, like nature of the particle, its surroundings, temperature etc., the probability distribution of the particle’s displacement (after a sufficient lapse of time) follows the Gaussian law of width $\sqrt{t}$.

Central Limit Theorem (CLT)

The CLT [75] states that the sum of a large number of independent, identically distributed random variables (i.i.d.r.v.), with finite variance, converges to a Gaussian distribution with the variance linearly proportional to the number of terms in the sum. In particular, let $x_1, x_2, x_3, ..., x_N$ be N (with $N \rightarrow \infty$) independent, identically distributed random variables,

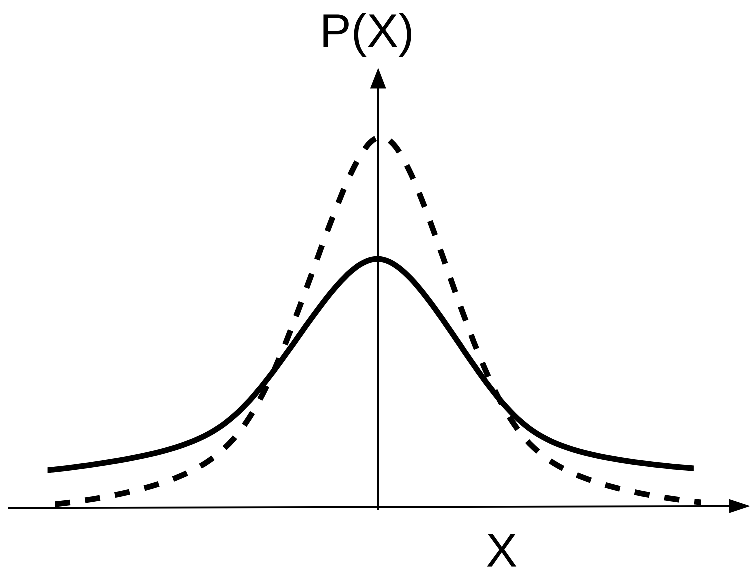

Figure 1.9: *Probability distribution P(X) of the total displacement X (which is the sum of large number of random displacements), where, the dotted and the solid curves give the Gaussian and the Lévy distributions, respectively. For large distributions, while the Gaussian distribution drops rapidly to zero, the Lévy distribution exhibits a fat/heavy tail (implying a finite probability of occurrence of large X).*

with their sum
$X_N = \sum_{i=1}^{N} x_i = x_1 + x_2 + ... + x_N : N \to \infty$.
Let, the mean of each of the i.i.d.r.v. be zero (without loss of generality) and variance be σ^2, then, CLT states that $p(X_N)$, the probability density function of X_N is given as :

$$p(X_N) = \frac{1}{\sqrt{2\pi\sigma_N^2}} exp(-x^2/2\sigma_N^2) \tag{1.4}$$

This is a Gaussian distribution with variance $\sigma_N^2 = N\sigma^2$.
In particular, for the normalized random variable, Z_N, corresponding to X_N defined as $Z_N = X_N/\sigma\sqrt{N}$, such that mean of $Z_N = 0$ and variance = 1, CLT states that probability density function of X_N (when properly normalized (Z_N), i.e., $p(Z_N)$) converges to a normal distribution or the Gaussian distribution with zero mean and unity variance.

$$p(Z_N) = \frac{1}{\sqrt{2\pi}} exp(-z^2/2) \equiv N(0,1) \tag{1.5}$$

The CLT is valid under very general conditions. Irrespective of the law of probability followed by x_i (: $i = 1, 2, ..., N$) and irrespective of their mutual correlations (provided these are weak), the probability distribution of X_N always remains a Gaussian curve, whose width increases as $\sqrt{N}$. Thus, the Gaussian acts as a sort of "black hole" of statistics and attracts and controls a vast majority of sums of random variables. In general, the total displacement of a Brownian particle is the sum X of the small, elementary random displacements x resulting from collisions with the surrounding molecules. The Gaussian law predicted by the CLT for this problem is almost always observed experimentally. The CLT, however, fails when the second moment of the distribution of the random variable ($< x^2 >$) is infinite.

1.4 Lévy statistics

Lévy flights

The condition for applying the classical CLT (i.e., finiteness of $< x^2 >$) is so frequently satisfied that its universality is implicitly believed. However, physical phenomena can exhibit statistical properties that are beyond the usual CLT. In such cases, CLT is violated and a non-Gaussian distribution is obtained, where the mean may or may not exist but the variance always diverges. These random walks termed as Lévy flights [76], have infinite mean-squared step size and arise for such random series of events where the rare or atypical events are so large in magnitude that they dominate the numerous small events. This results in a finite probability of observing improbable events giving the distribution a "Fat or Heavy tail" in the asymptotic limit (Fig. 1.9 : solid curve). Lévy distributions [77] are hence also termed as *"Fat or Heavy tail distributions"*. Further, the Lévy sums are exceptional in that they are rigorously "hierarchical". A very small number of terms dominate all others and the contribution of the latter to the sum X_N is negligible. Thus, in Lévy distributions, the determinant event is the rare event. and the sum reflects mainly the value of the largest terms. On the other hand, Gaussian sums are "democratic", with each term contributing significantly to the final result. A generic name for this phenomenon of rare intense events is intermittency.

In particular, probability density $f(x)$ of a random variable x with power-law tails :

$$f(x) \approx \frac{1}{x^{1+\alpha}} \qquad : \; for \;\; x \to \infty \tag{1.6}$$

(where α ensures normalizability) are simple laws that tend to appear frequently. If $\alpha \geq 2$, the variance $< x^2 >$ is finite and the usual or classical CLT applies. On the contrary, if $\alpha < 2$, i.e. the probability $f(x)$ of obtaining a given value of x decreases less rapidly than $1/x^3$ for large x, the variance diverges, x is said to have a "broad" probability density, and the usual CLT does not apply. If $\alpha \leq 1$, even the mean, $< x >$, diverges.

The first in-depth study of probability distribution with infinite moments was done by Pierre Paul Lévy. He was concerned with a random walk whose probability distribution for each jump has infinite moments. Consider an N-step random walk in one dimension, with each step of random length x governed by the same probability distribution $p(x)$, with zero mean. For such a N-step random walk Lévy investigated the circumstances in which the probability $P(X_N)$ for the sum of N steps $X_N = x_1 + x_2 + ... + x_N$ have the same distribution $p(x)$ (up to a scale factor) as the individual steps. Thus, the probability distribution for the position of the random walker after N steps is the same as that after one step (except for scale factors). This is basically the question of fractals, of when does the whole (distribution for N steps) look like its parts (the distribution for a single step). The standard answer is that $p(x)$ should be a Gaussian, because a sum of N Gaussians is again a Gaussian with a finite variance which is N times the variance of the original. But, Lévy proved that there exist other solutions such that $p(X_N)$ and $p(x)$ have the same distributions. All the other solutions, however, involve random variables with infinite variances. His answers, (most simply expressed in Fourier space) have the following form for the probability in the Fourier (k) space

$$p_N(k) = \int_{-\infty}^{\infty} p_N(x)\, exp(ikx)\, dx = exp\,(-constant \times N|k|^{\alpha}) \qquad : \; 0 < \alpha \leq 2 \tag{1.7}$$

Indeed, applying an inverse Fourier transform ($k \to x$) to Eq (1.7) gives

$$p_N(x) \sim constant \times N/|x|^{1+\alpha}, \qquad x \to \infty \tag{1.8}$$

$\alpha = 2$ is the Gaussian distribution. Cauchy distribution is the case $\alpha = 1$, which, when transformed back into real (x) space, has the form

$$p_N(x) = \frac{1}{\pi N} \frac{1}{1 + (x/N)^2} = \frac{1}{N} p(x/N) \tag{1.9}$$

which explicitly shows the connection between a one-step ($p(x)$) and an N-step distribution ($p_N(x)$). While for $0 < \alpha < 2$, variance diverges but mean is finite, for $0 < \alpha < 1$, both the mean and the variance are divergent. The power law for the tail of the distribution in Eq (1.6) indicates the absence of a characteristic size for the random walk jumps, unlike the Gaussian distribution ($\alpha = 2$). It is just this absence of a characteristic scale that makes Lévy flights scale-invariant fractals. Geometrically, this implies the fractal property that a trajectory, viewed at different resolutions, will look self-similar. Further, the sums governed by Eq (1.7) with $\alpha < 2$ are dominated by their largest terms, and thus by rare intermittent events and the power-law behaviour of the tail of the distribution function defines the appreciable probability of large values of displacement x, explaining the reason for Lévy flights. The exponent α will turn out to be the dimension of the point set visited by a Lévy flight. For Lévy flights this dimension is fractal ($0 < \alpha < 2$).

Generalized Central Limit Theorem (GCLT)

Paul Lévy, in the 1930s generalized the classical CLT to take into account the possibility of the existence of infinite moments, in particular divergent variance. He showed that for such cases, the sum X_N increases faster than the square root of the number of terms it contains, and the distribution obtained is no longer a Gaussian, but obeys a law which is now called the stable or Lévy-stable law.

In particular, Generalized Central Limit Theorem (GCLT) states that the sum X_N of large number of i.i.d.r.v., say $x_1, x_2, ...x_N$ converges to a Lévy-stable law. Thus, GCLT shows that if the assumption of finite variance is dropped, the only possible resulting non-trivial limits are Lévy-stable.

In fact, the interest of physics community for the GCLT seems to be stimulated by several arguments; first, the phenomena obeying only the GCLT, i.e. those with asymptotic power-laws for probability density ($f(x)$) with $\alpha < 2$, exhibit a statistical behaviour which is markedly different from the behaviour of the phenomena obeying the usual CLT. It is thus important to identify whether a physical process comes under the generalized form of the CLT, and then avoid the use of natural but irrelevant concepts, such as the average value, derived from the usual CLT. Second, the GCLT provides an efficient tool for the quantitative study of some physical problems, e.g. Lévy flight theory of laser cooling of atomic gases. In this case, it is worth noting that the ideas derived from the GCLT have had practical consequences, leading to more efficient cooling strategies and to record low temperatures. Finally, the GCLT also provides a useful qualitative insight for some random walks even when it is not strictly valid.

In the recent years, it has been increasingly recognized that the GCLT can help explain many physical processes, e.g., random walks in solutions of micelles [78], turbulent and chaotic transport [79, 80], diffusion of spectral lines in disordered solids [81], thermodynamics [82, 83, 84], granular flows [85], laser trapped ions [86, 87] and even fluctuations in the share prices.

Lévy walk

Inspite of the beauty and elegance of Lévy flights, the infinite moments of the distribution proved a stumbling block for its any meaningful use as a mathematical device to tackle physical problems. However, the divergence of the moments can be overcome by associating a velocity with each flight segment. One then looks at the displacement after a time t, which is a well-behaved time dependent moment of the probability distribution, rather than after N steps, which is infinity. This random walk with a velocity is called a Lévy walk or a Lévy drive (Fig. 1.10(b)) to distinguish it from a Lévy flight (Fig. 1.10(a)) where the walker visits only the endpoints of a jump and the notion of velocity does not arise. Thus, for a single jump in a Lévy flight the walker is only at the starting point and at the end point and never in between. In contrast to this, for a jump in a Lévy walk, the walker follows a continuous trajectory from its starting to its end points and hence a finite time is needed to complete the drive. The sites visited by the Lévy flight are the turning points of the Lévy walk. The mean square displacement for a jump in a Lévy flight is infinite and not useful. For a Lévy walk the random walker moves with a finite velocity (which depends on the jump distance) and hence its mean square displacement is never infinite but it is a time dependent quantity. Thus, a divergent result for the mean square displacement is avoided and replaced by a time-dependent result by associating a time scale with jump distances so that instantaneous jumps are not allowed and the long steps are penalized. Thus, in contrast to the Lévy flights, Lévy walks assume that a certain time is needed to complete each jump depending on its length. This aspect makes them more physical than Lévy flights and is the main reason for their widespread applicability.

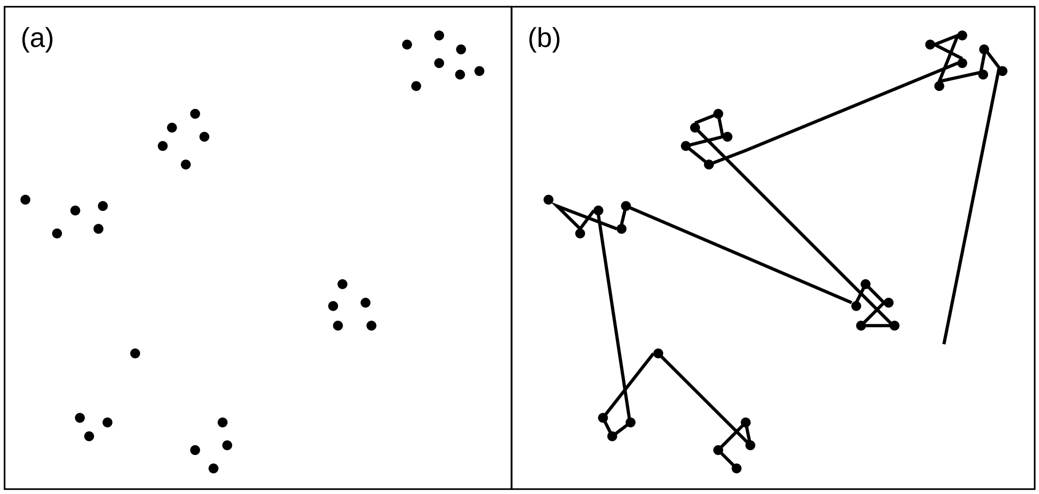

Figure 1.10: *Schematic of (a) Lévy flight, (b) Lévy walk. The Lévy walk includes the same set of points as the Lévy flight as well as the trajectory connecting these points. The Lévy flight points are turning points of the Lévy walk.*

The formulation of Lévy walks rests on a continuous time random walk (CTRW) approach where the emphasis is on the time rather than the number of steps. In the framework of CTRW [88, 89], for a Lévy walk, velocity is introduced through a coupled spatial and temporal probability $\Psi(r,t)$ for a random walker to undergo a displacement r in time t.

$$\Psi(r,t) = \Psi(t|r)\; p(r), \tag{1.10}$$

where, $p(r)$ is the probability function for a single jump. $p(r) \sim r^{-(1+\gamma)}$ and $\Psi(t|r)$ is the conditional probability that the jump takes a time t, given that its length is r. For simplicity, let

$$\Psi(t|r) = \delta\left(t - \frac{|r|}{v(r)}\right) \tag{1.11}$$

This ensures that $r = vt$. Lévy walks are thus a modification of the Lévy flights, preserving the spatial self-similarity.

Examples of Lévy flights/walks

Lévy flights were first used by physicists to explain experimental observations of photoconductivity in amorphous materials [90, 91]. Several studies since then have observed Lévy statistics in varied systems. Many physical examples of the Lévy statistics are known, e.g., anomalous diffusion in living polymers [78], strange kinetics [79], rotating fluid flow [80], subrecoil laser cooling [92], and interstellar scintillations [93]. Solid state physical example is in the context of spin-electronics [94, 95] (narrow metal-insulator-metal tunnel junction) giving large current fluctuations.

Lévy flights were first discovered experimentally by A. Ott and coworkers [78] while studying the diffusion of fluorescent molecules within an assembly of giant cetyl trimethyl ammonium bromide (CTAB) micelles in salted water. These micelles are long, flexible cylinders ($\sim$ 50 angstroms in diameter), made of surface active molecules and salts. Though these micelles resemble polymers, unlike polymers, they split and recombine continuously and at random - hence the name living polymers. Some of these cylinders are extremely long while others are much shorter such that the movement of each cylinder is hindered by the presence of others. Thus the longer a micelle, the slower is its movement. Every time a micelle splits and recombines with another one, the fluorescent molecule is found on a micelle whose size and hence the mobility is changed, i.e., the fluorescent molecule borrows a series of vehicles with varying performances. It was noted that it is the shortest micelle which made the fluorescent molecule travel the major part of the distance in one strike, the total contribution of other micelles being comparatively small. However, the probability of finding the tracer on a longer micelle is obviously greater. It was experimentally shown that the movement of the fluorescent molecules is exactly a Lévy flight.

Peng *et al.* analyzed the time intervals between heartbeats. They found that the erratic patterns observed in the heartbeats of healthy subjects can be described by Lévy distribution (α = 1.7) while data from patients with severe heart failure are much closer to a Gaussian distribution. They suggested that these results could arise from a nonlinear competition between branches of the involuntary nervous system.

Swinney *et al.* studied the flow of a liquid in a rotating vessel. The experimental set up essentially generates fluid flow in two dimensions. They found that vortices, a signature of turbulence, appeared at various places in the liquid. Tracer particles were followed for long periods of time and were found to alternate between staying in a particular vortex and flying towards a neighbouring one. The flights between vortices were found to follow a Lévy distribution (α = 1.3).

Oliveira *et al.* used both experimental and numerical methods to study a leaking tap. They found that the time intervals between drops fluctuate with a Lévy distribution (with α in the range 1.66-1.85).

Further, Mantegna showed that financial markets also can follow Lévy distributions. It was thus realized that this behaviour could provide a framework for developing econo physical models of share prices.

Researchers found that even the wandering albatrosses live their lives by a Lévy distribution. When looking for food, these seabirds fly for long distances, then forage in a small area before flying off again. Scientists are now investigating whether the foraging behaviour of other species, such as ants and bees also follow Lévy distributions. (It is similar to an airline making many local (national) short-distance flights followed by long-distance international flights).

1.5 Lognormal Distribution

Another distribution which resembles the Lévy distribution but is quite distinct from it is the lognormal distribution. Lognormal distribution is just the opposite of a typical Lévy distribution. In the latter, we essentially have a fluctuation-amplifier, e.g., an exponential barrier for quantum tunneling through or classical escape over that amplifies even a slight fluctuation in the barrier height. This is what gives a fat-tailed, broad Lévy distribution (as discussed in the previous section). Physically speaking, a lognormal distribution arises from the leveling property of the logarithm that deamplifies parametric fluctuations and hence gives a distribution which has all the moments finite, i.e., a light-tailed, narrow distribution [96]. More specifically, when the random quantity of interest (X) is a product of n random quantities ($x_i : i = 1, 2....n$) i.e., $X = \prod_{i=1}^{n} x_i$, (where x_i are i.i.d.r.v.), we have $ln(X) = \sum_{i=1}^{n} ln(x_i)$. Thus, the CLT should apply to $ln(X)$ and therefore $ln(X)$ should tend to a Gaussian limit as $n \to \infty$, i.e.,

$$p(X) = \frac{1}{\sqrt{2\pi\sigma^2}x}\, exp\left[-\frac{(lnx - \mu)^2}{2\sigma^2}\right], \quad x > 0 \tag{1.12}$$

where, μ and σ^2 are the mean and the variance parameters. Thus, in going from sum of i.i.d.r.v. to their product, one goes from the normal to the lognormal distribution.

It is to be noted that while, strictly speaking, the lognormal is light-tailed, narrow distribution in the asymptotic limit ($n \to \infty$), in the intermediate asymptotic limit (for n large, but not too large), it is effectively a fat-tailed, broad distribution.

To summarize, in this chapter we have provided the framework for discussing our original work done on RAMs, which form the subject matter of the remaining chapters of this Thesis.

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