Chapter 1

Introduction

In this thesis, we report our investigations on several aspects of wave propagation in disordered multiply scattering media, both passive as well as active. The study of wave transport in scattering media has a long and rich history since almost a century ago in the context of radiative transfer in the stellar atmospheres [1, 2, 3]. Also, the wave propagation and localization properties in spatially random media have been studied extensively for almost half a century[4, 5, 6, 7]. Now, in the modern context these are of importance in highly diverse areas such as mesoscopic systems, e.g. quantum wires, quantum dots and chaotic cavities [7], and medical imaging using laser light[8]. More recently, wave propagation and multiple scattering in the random amplifying media has assumed importance in the last decade, and its study has led to a new class of lasing media and random lasers. Inasmuch as it would be impossible to review all the previous work, we will attempt in this Chapter to provide a self-contained introduction pertinent to the work reported in this thesis. The work in the thesis can be classified broadly into two parts: one, concerning coherent wave transport in one-dimensional disordered systems, and the other concerning the development of stochastic models to describe the wave propagation in terms of an incoherent energy transport. An overview of the thesis is given in the preface of the thesis.

1.1   Scattering systems

Since most of the work in this thesis pertains to scattering, we will begin with a brief description of the scattering process. By scattering we mean a change in the energy and/or momentum of an incident particle, caused by a (change in the) potential.
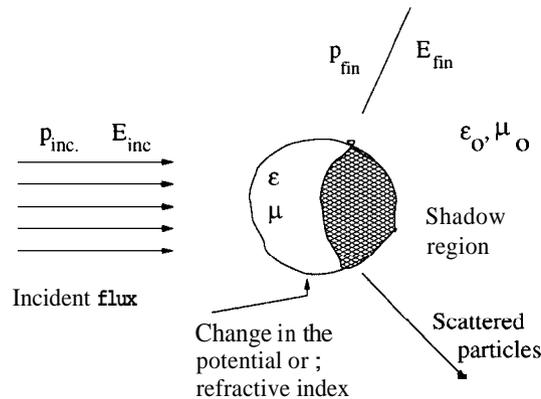
Figure 1.1: Schematic depiction of scattering of a flux of particles incident on a fixed scatterer, resulting in changes in the energies($E_{inc} \rightarrow E_{fin}$) and the momenta $(\vec{p}_{inc} \rightarrow \vec{p}_{fin})$ of the incident particles. The scatterer in this case is assumed to be a 'hard' body. The shadow region for a classical particle is indicated by the shaded region.

The particle is taken to be asymptotically free, initially as well as finally. In the case of a quantum particle or a wave, this will also be accompanied by a change in the phase of the wave-function, which needless to say, does not have a classical particulate analogue. These changes in energy, momentum and phase are governed by the conservation laws for energy and momentum, and by the nature of the scatterer. A notable difference between the classical and the quantum cases is that there is a "shadow region" for the classical case which is not there for a wave because of diffraction. This is schematically depicted in Fig. 1.1, where scattering from a 'hard'-body is considered: the shaded region is inaccessible for a classical particle. Any change in the shape of the potential in that region would not be felt by a classical particle, while the scattering of a quantum particle or wave will be affected. The scattering process can be elastic, where the kinetic energy is preserved, or inelastic where there is exchange of kinetic energy with some internal degree of freedom (e.g. atomic/molecular excitations) or with an external system (e.g. phonons in an atomic lattice or other electrons). In the case of quenched disordered systems that we will consider here, the scattering will always be elastic. Inelastic scattering events such as electron-phonon or electron-electron scattering lead to dephasing of the electron wave, *i.e.*, disappearance of interference of the complex wave amplitudes following alternative paths. In contrast elastic scattering by static disorder, however strong,

cannot cause dephasing.

The strength of scattering from a scatterer is characterized by the differential scattering cross-section which is the ratio of the intensity scattered in a given direction per unit solid angle to the incident intensity. The total scattering cross-section(σ_s) is the total scattered intensity over all directions, and is given by the integral of the differential scattering cross-section integrated over all directions. Although the scattering cross-section are defined in terms of intensity, the wave interference affects them and as such must be taken into account to compute the cross-sections.

In the following, we will explicitly talk about light scattering (as a matter of personal preference), though the general philosophy is applicable to all kinds of waves: electrons, neutrons and classical waves such as light and sound. One point that has to be kept in mind is that light is a vector wave, *i.e.,* it has a polarization transverse to the direction of propagation. The polarization properties are, however, ignored for most purposes in this thesis and the treatment is for a scalar wave. Scattering of light is caused by a spatial change in the refractive index of the medium. One can. of course, also consider the cases of reflection and transmission as special cases of scattering.

1.1.1   A single scatterer

Consider the scattering of a light wave by an isolated scatterer. The description of scattering and absorption of light by even a single small particle is quite involved. For details, the reader is referred to Refs. [9, 10]. It suffices for our purposes to note that there are three different regimes of scattering depending on the length-scales in the problem, namely, the wavelength (A) of the incident light and the size of the scatterer (a).

For a small scatterer (a $\ll$ A), the entire scatterer gets uniformly polarized by the incident light, and acts as a single dipole. When it re-radiates, the scattered field is that of the dipole radiation - this is the well known Rayleigh scattering. The scattered intensity is isotropic in the plane perpendicular to the polarization of the incident light and has a $\cos^2 0$ dependence (0 being the angle of scattering - see Fig. 1.2a) in the plane of polarization. It also has a λ^{-4} dependence on the wavelength, implying that the shorter wavelengths are scattered more efficiently. This was used to explain
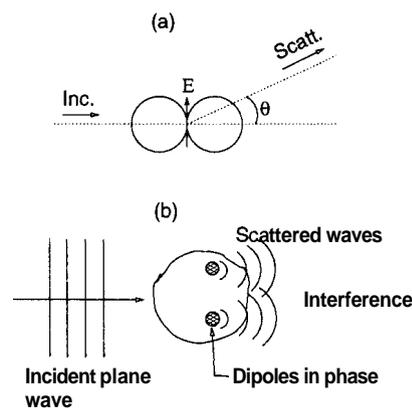
Figure 1.2: (a) Schematic depiction of Rayleigh scattering for a wave with polarization in the plane of scattering. (b) Interference of waves re-emitted by different parts of a scatterer resulting in an angle dependent scattering pattern

the blue colour of the sky as due to the scattering of sunlight by air molecules, the blue light being scattered about five times more strongly than the red. When the size of the scatterer is comparable to the wavelength of the incident light (a $\sim$ A), then the entire particle no longer behaves as a single dipole. Different parts of the inhomogeneously polarized particle re-emit coherently, but now with different phases and we get an interference between these re-radiated waves. This is shown schematically in Fig. 1.2b. The coherence and the mostly constructive interference cause a great increase in the scattering cross-section. This is the reason for the water droplets in clouds to strongly scatter light. If there are N-dipoles oscillating coherently within the particle of size NA, then the scattering cross-section increases as N^2!!!Of course, increasing the size of the particle to sizes much greater than a wavelength does not help because then there is as much destructive interference as constructive on an average. It might so happen that the constructive and destructive interferences might choose to channel the light into some direction(as in reflection) due to the morphology of the situation. In the limit a $\sim$ A, the scattering properties of the small particle are quite complex and one needs a full solution of the wave equation for the system. Thus, we have the Mie theory, where analytical solutions for spheres, spheroids and cylinders for an incident plane wave have been obtained [9]. The salient features of these solutions are: (i) at small sizes or low frequencies, we get the Rayleigh scattering behaviour, (ii) at intermediate sizes (a $\sim$ A), the scattering cross-section increases and passes through a

number of peaks - the Mie resonances where the scattering cross-section can become as large as six times the geometric cross-section, and (iii) the high frequency or the large size limit (a $\gg$ A) where the cross-section saturates to the geometric optics limit of $2\pi a^2$ which is twice the geometric area, an effect caused by the wave nature of light (the scattering due to the shadow region). This is the third macroscopic regime - in the limit of (L $\gg$ A), when the geometric optics can be applied and the scattering can be considered as reflection from a dielectric interface/discontinuity of linear dimension L. Another notable feature is that the scattering which is nearly isotropic for small scatterers (a $\ll$ A), scatters more in the forward direction as the scatterer size is increased (a $\sim$ A), and for very large scatterers, almost all the light is scattered in the forward direction. This is due to the fact that the light scattered by N independent scatterers always interferes constructively in the forward direction due to the symmetry of the problem [9]. This forward scattering behaviour will have to be incorporated in any realistic theory. A useful parameter which captures the essence of this forward scattering nature is the anisotropy factor g which is defined as the average of $\cos\theta$ (θ being the angle of scattering) over all the angles for a given scatterer.

1.1.2 Collection of scatterers: Single scattering and multiple scattering

Now let us consider what happens to a wave as it enters a medium containing randomly placed scatterers. On a gross macroscopic level, it is treacherously simple. The incoming beam gets attenuated due to the scattering out of the incident beam and a diffuse glow emanates from the medium. On the microscopic level, however, the situation is extremely complicated. The light could be scattered thousands of times before leaving the sample. It is clear that there are two limiting scattering regimes. One, the single scattering regime, when the scatterers are weakly scattering and the density of the scatterers is also small, so that the probability of the light which is scattered once to be scattered again before leaving the scattering region by other scatterers is extremely small. In this case, the mean free path of the light is much larger than the sample size and the scattering acts only as small perturbation on the incident light'. The total scattered wave amplitude in any direction is given by the sum of the scattered amplitudes by the individual scatterers, i.e., $E_{tot}^s(\vec{r},\theta) = \sum_i E_i^s(\vec{r},\theta)$. In the

case of the positions of the scatterers being random, the phases of the waves scattered by different scatterers would be random, and so the intensities would add up. This is true in all directions except the forward scattering direction, where all the scattered waves interfere constructively. The scattered intensity in different directions (angular scattering profile) would be N times that of a single scatterer (if the N scatterers are identical). As the density of the scatterers increases, we can incorporate the first order corrections in this single scattering picture, by considering that the incident wave gets attenuated by scattering (and absorption) as it propagates into the medium.

The second regime is that of strong scatterers present with high density so that the light is multiply re-scattered many times before finally emerging. In this multiple scattering regime, the scattering is a very strong perturbation on the incident light field and the wave amplitude at any given scatterer is strongly modified by the presence of other scatterers. In this situation, two problems arise. First, we do not know the incident field on a given scatterer, and second, we know the scattering cross-sections only for a plane incident wave (that too only for simple shapes of the scatterers). In fact, the near-field wave amplitudes can vary in a very complex manner. Now, we realize that we are faced with the problem of solving the full wave equation over the entire configuration of the scatterers. However, we are not interested in a particular configuration, but want ensemble averaged behaviour over all the possible configurations. Thus we realize that the detailed information is quite unnecessary and only quantities such as the average cross-sections and the average density of scatterers would matter. Hence, we define characteristic length scales for the problem, i.e., the scattering mean free path $l = \langle (\rho \sigma_s)^{-1} \rangle$ and the transport mean free path $l^* = l/(1 - g)$, where p is the density of the scatterers, a, is the scattering cross-section and g is the average anisotropy factor for the scatterers. The transport mean free path accounts for the forward scattering nature of the scatterers. Similiarly, we can also define the attenuation, or gain length, depending on the absorption, or the gain, in the medium.

We are faced here with two possibilities to proceed further. One, we can carry on our studies in the multiple scattering domain by considering the wave propagation with all the interference effects. This is the multiple scattering wave theory resulting in the Twersky-Foldy integral equations for the wave amplitude and the correlation

function[11] and are equivalent to the Dyson and the Bethe-Salpeter equations for the single and the double Green functions[12]. Obviously, it is not simple to solve these equations, and approximations are made in a systematic perturbation theory by considering only scattering events of a particular class and summing the perturbation series of that class to all (infinite) orders. Only in one-dimensional problems has a non-perturbative treatment been possible. The second approach is much simpler. Here we proceed on the assumption that interference effects may not be dominant, and treat the whole problem heuristically by considering only the transport of energy, or particle flux subject to conservation. It turns out that this approach fails in the limit of strong scattering($kl^* \sim 1$) when interference effects become important. We will be using both these approaches in this thesis, within their respective domains of validity and utility.

1.2   Random media, stochastic media and models of disorder

Before we proceed further, we will note that the randomness in the potential or the refractive index can be of two kinds. In the first case, the potential is static in time, but can vary in space, the variation being random. This is the case of quenched disorder and in this thesis, we will call this kind of media as random or disordered media. In this case, the interference effects will never really be washed away, although the phase of the wave propagating in the medium can vary randomly in space. The other case is that of temporally random media, or stochastic media, where the randomness of the potential is not in space but in time - the scatterer strengths or positions vary in time in a random manner. In this case, the interference effects are not important (See Section 1.3.1). The important difference between these two cases is that, if a wave returns to its initial position after some scattering events, it feels the same scattering potential in the former case of spatial randomness, but a different potential in the latter case of a stochastic medium (as the potential has changed over that time). This has important implications for wave interference and coherent backscattering (See Sections 1.3.1 and 1.4.1). In the next three chapters of the thesis (Part A), we will consider the case of spatial randomness and the associated wave interference effects. The treatment in the remaining three chapters (Part B)

is applicable to stochastic media, where we consider only an energy (particle flux) transport.

To describe the randomness of the potentials mathematically, we need to have rnodels for the randomness which we present in the following. The simplest model is to assume an Gaussian white *uncorrelated* noise: If $V(x)$ be the potential as a function of the co-ordinate x, then we have $\langle V(x) \rangle = 0$, $(V(x)V(x^1)) = V_0^2 \delta(x - x')$. All the odd moments vanish and the higher even moments can be expressed in terms of the second moment due to the Gaussian nature of the fluctuations about the mean[13]. This model is simple to treat mathematically and has been used extensively. The next step is to recognize that in no real case would the potentials be completely uncorrelated, and to consider a correlated random potential with a finite correlation length. The mathematical treatment becomes much more complex, and only exponentially correlated randomness has been amenable to mathematical treatment so far, i.e., for $\langle V(x)V(x') \rangle = V_0^2 \exp(-\Gamma |x - x'|)$, where Γ^{-1} is the correlation length. In this context, the Telegraph disorder or the dichotomic Markov process, is important. Here the random variable can assume only two values and is exponentially correlated. In this thesis, we have used the Gaussian White-noise and Telegraph-disorder models to describe the randomness. As to the relevance to real optical scattering media, the continuum Gaussian white-noise would describe well a scattering intralipid suspension or a biological tissue where the correlation is quite minimal. On the other hand, a telegraph noise model would be apt for a colloidal suspension of monodispersed microspheres. In the electronic case of scattering of Bloch waves by atomic impurities, either the strength or the position of the scattering potential can be treated as random corresponding respectively to the substitutional disorder or the topological disorder. In Chapter-4, where we describe a one-dimensional chain by a tight binding Hamiltonian, we have used a random site energy uniformly distributed within a given range - the uniform diagonal disorder model. Other models of disorder are also possible, but the Gaussian white-noise model remains the clear favourite of researchers in view of its mathematical simplicity.

1.3 Wave propagation, diffusion and energy transport

In the study of wave propagation in a multiply scattering medium, one must necessarily distinguish between two cases, vix., weakly scattering media and strongly scattering media. In the former case of weak scattering, the mean free path of the wave in the medium is much greater than the wavelength, and wave interference effects are found to be negligible in this regime. The entire phenomenon can then be described as a gross energy transport by classical diffusion. In the latter case of strongly scattering media, there are important effects of wave interference on the wave propagation, and this can even result in a total localization of the wave. In this Section, we will present a brief introduction to the transport of wave energy in weakly, but multiply, scattering media. The case of coherent wave transport in strongly scattering media will be discussed in the next Section.

In the modern context, the description of wave propagation in weakly scattering random media as an incoherent (where wave interferences are neglected in a sense described in Section 1.3.1) energy transport has found important applications in biomedical imaging and therapy. This is, of course, apart from its traditional applications in astrophysics[2], atmospheric sciences and remote sensing [3]. The use of light and lasers in medicine have increased manifold in the past decade. Enormous efforts have been devoted to the development of new diagnostic techniques such as near-infra-red imaging [8] and flourescence spectroscopy of tissues [14], as well as therapeutic uses such as photothermal coagulation and Photo-dynamic therapy [15]. Tissues are highly scattering media and an accurate knowledge of light transport is indispensible to developing more accurate diagnostic techniques using light. The scattering by biological media is, however, weak and a description in terms of an incoherent energy transport suffices for most purposes. This is also called the photon migration problem in turbid media. Another context, where such a description is applicable, is the random diffusion laser [16] i.e., dense, weak scatterers randomly imbedded in an amplifying medium. We will deal with the latter case in Section 1.5.1 in more detail.

In the description in terms of an incoherent energy transport, light is treated as particles (photons) bouncing off randomly placed scatterers with the coefficient

of restitution being unity, *i.e.*, elastic scattering (absorption removes particles while amplification adds more particles). Now, these particles execute a random walk in the medium. Conservation of energy (number of particles), yields a Boltzmann-like transport equation (presented in Section-1.3.2). As can be expected, the random walk problem is accurately described by the diffusion equation at length scales much larger than the mean free path of the wave$(L \gg l^*)$. This has resulted in the diffusion equation becoming the work-horse of the biomedical community. However, at smaller length scales, the diffusion picture becomes inaccurate. Recently there has been considerable interest in the description of multiple light scattering at small length scales (L $\sim l^*$) and small time scales *(t $\sim$ t*, where t* is the transport mean free time)*, both from the point of fundamental physics [17] and from the point of medical imaging, where the early arriving 'snake' photons are used to image through human tissues [18, 19]. It has been experimentally shown that the diffusion approximation fails to describe phenomena at distances of L < 8l* [20]. Moreover, the diffusion approximation, which is strictly a Wiener process for the spatial co-ordinates of a particle, is physically unrealistic. This is the motivation for our attempt to develop more realistic models of photon migration presented in the last three chapters (Part-B) of the thesis.

1.3.1   Random phases: connection between wave propagation and diffusion

One knows that the wave propagation is strictly governed by the Schrodinger equation for the quantum electron waves and by the Maxwell's equations (or the Helmholtz equation) for the case of light (classical waves). On the other hand, we do know that the energy transport in a random medium at a macroscopic level is to a large extent diffusive, and hence, described by the diffusion equation. The main problem is to reconcile the final irreversible diffusive dynamics of a system propagating under the completely reversible wave equation. The loss of coherence and phase memory in such a random system is not completely understood yet.

In a multiply scattering random medium, there are infinitely many possible paths for a wave to propagate. The total probability amplitude for the wave to propagate from one point to another is given by the sum of the probability amplitudes of these waves for traversing all these allowed paths. Consider the partial waves traversing
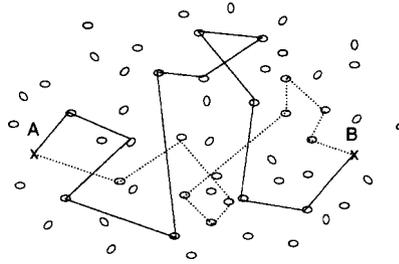
Figure 1.3: Two possible paths for a wave multiply scattered in a random medium.

through two such paths shown in Fig. 1.3 from A to B. The amplitude at B is given by the sum of the amplitudes for the partial waves going through the two paths: $\psi(B) = \psi_1 + \psi_2$ and the intensity is proportional to

$$|\psi(B)|^2 = |\psi_1|^2 + |\psi_1|^2 + 2|\psi_1\psi_2|\cos(\phi_1 - \phi_2), \tag{1.1}$$

where $\phi_{1,2}$ are the phases of the two interfering partial waves. Now, the relative phase of the interfering partial waves will vary randomly with the choice of the random paths. For any given configuration of the *spatially* random medium we end up with a random interference (speckle) pattern, *i.e.*, at some points the partial waves propagating through all the possible paths in that configuration add up constructively and at some others destructively. The spatially quenched disorder will always result in such a speckle pattern which cannot be washed away by any amount of static disorder. These are characteristic fluctuations of the random medium. For the ensemble average of all possible configurations of the random medium, however, the interference terms are expected to cancel out on an average and we would expect that the probabilities (intensities) to add on too. This, indeed, turns out to be the case for weakly scattering media in higher dimensions and such a behaviour is called self-averaging behaviour (See Section-1.7). Such an averaging can be accomplished temporally, for e.g., by Brownian motion of the scatterers in a colloidal suspension of microparticles.

We will now define characteristic *coherence lengths* of the wave in the random medium: the *longitudinal* coherence length and the *transverse* coherence length. The longitudinal coherence length is related to the fact the coherence is broken by a stochastic process in time and after a certain coherence length, the phase of the wave changes suddenly. For the case of electron, this would correspond to the inelastic

mean free path, a limit set by the inelastic scattering processes and for the case of light, it would be the coherence length of the laser or the light source itself. The transverse coherence length comes about because of the spatially random phases of the wave at different spatial points caused by random scattering events. This acts effectively as an incoherent (or partially coherent) source for propagation to another point. Thus, we have *coherence bundles of paths* (if they originate from within a transverse coherence length) where interference is important. If the problem involves transport through many different coherence bundles of paths, then the interferences would be expected to effectively wash out and an incoherent energy transport results. Obviously if very long paths (much longer than the longitudinal coherence lengths) are important for the transport, then again an incoherent transport of energy results (e.g. Drude conductivity in a metal).

For the case of dynamic (time-dependent) disorder when the potentials $V(\vec{r}, t)$ vary randomly in time, the Schrodinger equation describing wave propagation

$$i\hbar \frac{\partial \psi(\vec{r}, t)}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \psi(\vec{r}, t) + V(\vec{r}, t) \psi(\vec{r}, t) \tag{1.2}$$

can indeed be reduced to a diffusion equation for the probability density. Writing the wave function $\psi = \sqrt{\rho} \exp(i\theta)$, and substituting into the Schrodinger equation, we obtain

$$\frac{\partial \rho}{\partial t} + \frac{\hbar}{m} \vec{\nabla} \cdot (\rho \vec{\nabla}\theta) = 0. \tag{1.3}$$

Now, we can take $6'$ to be randomly varying in time due to the inelastic scattering processes such as electron-phonon scattering etc. Specifically, for the case of $\vec{\nabla}\theta$ being a white noise, the above equation reduces to a diffusion equation for the average quantity (ρ):[1]

$$\frac{\partial \langle \rho \rangle}{\partial t} = \frac{\hbar^2 \Delta^2}{2m^2} \nabla^2 \langle \rho \rangle, \tag{1.4}$$

where $(\ )$ indicates averaging over the stochastic process and Δ is the r.m.s. fluctuation in $|\nabla\theta|$.

In the case of purely static disorder, however, it has not yet been satisfactorily understood as to how the diffusive solutions can be obtained from the wave equations. The diffusion equation is recovered as an approximation when only a subclass of the multiple scattering terms, i.e., those corresponding to the ladder terms are taken

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[1]Here use of the Novikov's theorem has been made – see Appendix B.

into account[7, 12, 21]. The radiative transfer equation for the specific intensity can be obtained from the wave equation for the mutual coherence function(two-point correlation function), if wave fluctuations at small scales are neglected. The mutual coherence function is approximately related to the specific intensity through a Fourier transform. For very highly scattering media, and at large length scales, the radiative transfer equation goes over to the diffusion equation (See Section 1.3.4).

1.3.2 Radiative transfer and diffusion

As we have seen above, the randomization of the phase of the wave in weakly, but multiply scattering media enables us to describe the entire problem in terms of an incoherent energy transport. Historically, the latter approach was developed much earlier in the context of astrophysics as a radiative transfer theory for light propagating through stellar atmospheres[1, 2]. One of the most basic quantities needed to characterize the energy transport is the *specific intensity* $I(\vec{r}, \hat{\Omega}, t)$, defined as the average power flux density within a unit frequency band at a frequency ν within a solid angle for a given direction $\hat{\Omega}$ at any point Fin the medium. Mathematically,

$$dP = I(\vec{r}, \hat{\Omega}, t)\hat{\Omega} \cdot d\vec{a} \; d\Omega \; d\nu \tag{1.5}$$

is the power(dP) flowing within a solid angle $d\Omega$, through an elementary area $d\vec{a}$ oriented along the direction of the unit vector $\hat{\Omega}$, and in the frequency interval $(\text{v}, \nu + \text{dv})$. The specific intensity has also been variously termed the brightness or radiance in literature. Two other important quantities are the average intensity defined as $U(\vec{r}, t) = 1/4\pi \int_{4\pi} I(\vec{r}, \hat{\Omega}, t) d\Omega$, and the average flux density defined as $\vec{F}(\vec{r}, t) = 1/4\pi \int_{4\pi} I(\vec{r}, \hat{\Omega}, t)\hat{\Omega} d\Omega$,

The radiative transfer equation is essentially a statement about the conservation of energy flowing through the random medium and is a Boltzmann like transport equation:

$$\frac{\partial I(\vec{r}, \hat{\Omega}, t)}{\partial t} + c\hat{\Omega} \cdot \vec{\nabla} I(\vec{r}, \hat{\Omega}, t) = -\rho\sigma_t I(\vec{r}, \hat{\Omega}, t) + \frac{\rho\sigma_t}{4\pi} \int p(\hat{\Omega}, \hat{\Omega}') I(\vec{r}, \hat{\Omega}', t) d\Omega'$$
$$+ \; \epsilon(\vec{r}, \hat{\Omega}', t), \tag{1.6}$$

where ρ is the density of scatterers, σ_t is the total scattering cross-section defined as a, $+$ a,, a, is the scattering cross-section, a, is the absorption cross-section,

$\epsilon(\vec{r}, \hat{\Omega}', t)$ is the strength of the additional sources in the medium and $p(\hat{\Omega}, \hat{\Omega}') = 4\pi/\sigma_t \left| f(\hat{\Omega}, \hat{\Omega}') \right|^2$ is the phase function, $\left| f(\hat{\Omega}, \hat{\Omega}') \right|^2$ is the differential cross-section and $1/4\pi \int_{4\pi} d\hat{\Omega}' \, p(\hat{\Omega}, \hat{\Omega}') = \frac{\sigma_s}{\sigma_t} = W$, the albedo.

The main disadvantage of the above formulation is that the Radiative transfer equations have not been solved analytically even for the simplest geometry. Hence, in most treatments of the problem, one resorts to the *diffusion approximation*. In the diffusion approximation, it is assumed that the light has encountered sufficiently many scatterers and as a result of many random scattering events, the angular distribution of the specific intensity is *almost* uniform. The angular dependence cannot be absolutely constant, because then the average flux density would be zero and there would be no preferred direction of the net flow. Thus, the specific intensity is assumed to be Quasi-Isotropic:

$$I(\vec{r}, \hat{\Omega}, t) \simeq U(\vec{r}, t) + \frac{3}{4\pi} \vec{F}(\vec{r}, t) \cdot \hat{\Omega} \, . \tag{1.7}$$

Further, we assume that the time variation of the flux vector is slow compared to the mean free time, i.e, $|\partial \vec{F}/\partial t| \ll |\rho \sigma_t \vec{F}|$, and that the scattering phase function depends only on the cosine of the scattering angle. Then, we obtain the following diffusion equation [11]

$$\frac{\partial U(\vec{r}, t)}{\partial t} - D \nabla^2 U(\vec{r}, t) = \epsilon(\vec{r}, t) \tag{1.8}$$

where $D = cl^*/3$ is the diffusion coefficient, $l^* = 1/\rho\sigma_s(1 - g)$ is the transport mean free path in the medium and $\epsilon(\vec{r}, t)$ represents the diffuse sources or sinks in the medium. Absorption or amplification can be included in the source term as $\epsilon(\vec{r}, t) = c\sigma_a U(\vec{r}, t)$, where a, is the effective absorption/amplification coefficient. a, is negative for absorption and positive for amplification. The solution of the diffusion equation in an unbounded medium is a Gaussian in space, which expands in time. The absorption(amplification) causes an exponential decay(increase) in time.

The diffusion approximation has been utilized to describe phenomenologically, transport theory in numerous contexts in physics, chemistry and biology. It describes an irreversible process of evolution whereby any singularities are smoothened out and eventually a uniform background will prevail.

1.3.3 Inadequacy of the diffusion approximation

The diffusion approximation is valid for multiply scattering media with small absorption (gain) at points far away from any source (L ≫ I*). Similarly, for a pulse of light incident on the medium, the diffusion approximation becomes valid only at times much greater than the mean free time ($t \gg t^* = l^*/c$). The crucial assumption in the diffusion theory is that the specific intensity is quasi-isotropic. As can be expected, this isotropization is bound to take a few scattering events and will not be valid in a region of space close to the source.

One of the most striking experiments to verify the length-scales and time-scales at which the diffusion approximation is valid, was performed by Yoo and Alfano[20]. In this experiment the time-dependent transmission of a femtosecond pulse through a slab of disordered medium (a colloidal suspension of polystyrene microspheres) was directly measured by a streak camera, for several concentrations of the colloid. It was found that for slab thicknesses smaller than $10l^*$, the measured light arrived earlier that that predicted by diffusion theory. Clearly, this is an effect of the ballistic aspect of transport, which the diffusion theory does not account for.

The diffusion approximation is strictly speaking a Wiener process for the spatial position of the particle. The stochastic Langevin equation for the velocity of a particle undergoing pure diffusion is $\dot{\vec{r}}(t) = \vec{f}(t)$, where $f_i(t)$ are independent, Gaussian, white noise random variables and $f_i(t)$ are independent of $r_i(\tau)$ for $\tau \leq t$ (The Markovian property). This process is unphysical in that, the paths are non-differentiable almost everywhere and the velocity is undefined. Strictly, the diffusion approximation holds in the limit: the mean free path $l^* \to 0$, the speed (group velocity) $c \to \infty$ such that the diffusion coefficient $D = cl^*/3$ is a constant. In other words, the particle scatters at every point in space. Thus, the diffusion approximation neither accounts for a finite mean free path nor the constant speed of the wave in between the scattering events. Essentially, the pure diffusion process has no directional memory *i.e.,* it has zero persistence in the velocity space. In reality, the directional persistence is non-zero and the transport occurs in the phase-space. The stochastic process describing the spatial position is expected to have a long persistence citemajumdar due to the non-Markovian nature. Essentially, for the diffusion approximation to hold, we need to coarse grain the description over length scales much greater than the characteristic

length scales in the problem, *i.e.*, the transport mean free path (similiarly for the time-scales).

Thus, there is a need to have a more accurate, alternative model to describe photon migration at length scales and timescales comparable to the mean free path and the mean free time respectively, particularly for medical imaging purposes where the ballistic and snake (near ballistic) light is used. This lack of a simple and yet comprehensive model to address the quasi-ballistic regime has led to several *ad-hoc* proposals involving parametrization of the coefficients and the boundary conditions in the diffusion theory. For example, there has been much debate on whether the diffusion coefficient should depend on the levels of absorption or not [23]. The diffusion coefficient by the very definition should depend only on the scattering properties and absorption should be taken into account separately as another parameter in any theory.

1.3.4 Connection between the specific intensity and the mutual coherence function

In the radiative transfer theory, interference effects arising from multiple scattering are neglected. Only the interference/diffraction effects in each scattering event is accounted for explicitly by a rigorous wave theory calculation of the phase function used in the equation. Now the question is, do we lose all information about the interferences in the approximate radiative transfer equation? It actually turns out that much information concerning the field quantities is included in the radiative transfer equation. The mutual coherence function or the two-point correlation function $\Gamma(\vec{r_1}, \vec{r_2})$ is defined as

$$\Gamma(\vec{r_1}, \vec{r_2}) = \langle E(\vec{r_1}) E^*(\vec{r_2}) \rangle = \Gamma(\vec{r}, \Delta \vec{r}) \qquad (1.9)$$

where $\vec{r} = 1/2(\vec{r_1} + \vec{r_2})$, $\Delta \vec{r} = 1/2(\vec{r_1} - \vec{r_2})$, E is the wave amplitude and $\langle\ \rangle$ denotes the averaging over the disorder. When the correlation function is a slowly varying function of $\vec{r}$ (in space), *i.e.,* when wave fluctuations on a small length scale can be neglected, the mutual coherence function can be written as the Fourier transform of the specific intensity:[24, 25]

$$\Gamma(\vec{r}, \Delta \vec{r}) \simeq \int I(\vec{r}, \hat{\Omega}) \exp(iK_r \hat{\Omega} \cdot \Delta \vec{r})\ d\Omega \qquad (1.10)$$

where $K_r = \Re[k + 2\pi f(O\hat{O})/k]$ and $f(O O)$ is the complex differential backscattering coefficient. Using the above and the coherent potential approximation (CPA) for the single Green's function[7], the Twersky's integral equations for the intensity [11] can be shown to reduce to the integral form of the radiative transfer equation(1.6) for the average energy density.

Thus, we see that some information about the wave quantities, is indeed preserved in the radiative transfer theory. In fact, the above is equivalent to only including the ladder terms in the Bethe-Salpeter equations for the intensity (double) Green's function [12]. Basically, the phase space in the radiative transfer theory consists of $\vec{r}$ and $\hat{\Omega}$, which are essentially Fourier conjugate variables. In wave theory, the $\vec{r}$ and $\hat{\Omega}$ cannot be specified simultaneously. By analogy with quantum theory, this makes the specific intensity a non-measurable quantity [26]. There is an inherent ambiguity of the energy flux vector in the near field of a source or scatterer. The specific intensity, if defined, as the Fourier transform of the mutual coherence function, can turn out to be even negative in the near field zone[27], and does not possess all the properties of the specific intensity defined in conventional radiative transfer theory. Thus, the above should only be taken in an approximate sense, whereby the wave fluctuations at small length scales have been smoothened in a coarse-grained description. Specifying the direction of the energy flux intuitively in RTE seems to capture some of the coherence properties. It should be noted that the simple radiometric calculations can be considerably accurate and very effective to describe even coherence propagation with partially coherent sources, where rigorous wave calculations are difficult to carry out [28]. In our context, the simplification that the radiative transfer theory offers over the multiple scattering wave theory, can be exploited to incorporate as many wave properties as possible in a simple radiometric calculation.

1.4 Wave interference and Anderson localization

1.4.1 Coherent backscattering

In this Section, we will consider the case of wave propagation in strongly scattering media with quenched disorder, where we cannot neglect the interference effects. All the partial waves traveling through different paths in the medium will, of course, interfere with each other. This will result in a random but static speckle pattern, which
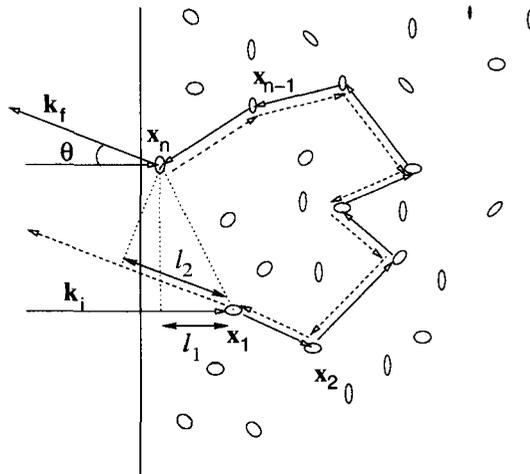
Figure *1.4:* A typical path in real space which constructively interferes with its time-reversed counterpart in the backward direction. Both the paths have the same incident and exit wave-vectors. The net path length difference is $l_2 - l_1$, corresponding to a phase difference of $(k_f - k_i) \cdot (x, - x_1)$.

will change from direction to direction and from sample to sample, as the phases of these partial waves change randomly from path to path. The coherent backscattering (CBS) effect arises from the constructive interference of any partial wave with its time-reversed counterpart in the medium. In the exactly backscattered direction, both these two partial wave amplitudes have the same phase and constructive interference results. The partial wave traversing the backward path has the same path length and scatters through the same set of scatterers as the forward path, though in the reverse order. (see Fig. *1.4).*

For the case of light this shows up as an enhancement of the backscattered intensity, with the intensity in the backward direction being twice than the intensity in directions far away (enhancement factor of *2)*[29]. Away from the backscattered direction, the counterpropagating paths develop a phase difference depending on the relative positions(separation $\sim l^*$) of the first and last scattering events in the medium. For the ensemble of all possible light paths, these phases will randomize and the reflection is enhanced within a narrow cone in the backward direction with an angular width of the order of λ/l^*. This peak shows up only after the ensemble averaging over the large scale sample specific fluctuations (speckle) that originate from the random medium [30]. An experimental measurement of CBS from milk is shown in Fig. 1.5.
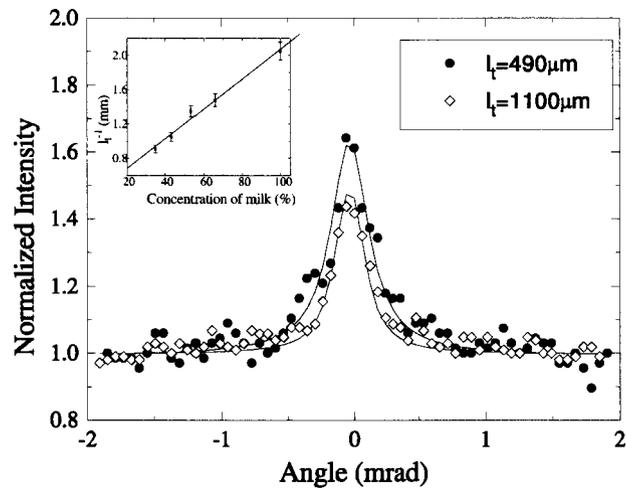
Figure 1.5: The CBS peak seen in the backscattering direction for light scattering from milk. The enhancement factor is lesser than 2 because of the finite resolution of the experimental setup. [Taken from S. Anantha Ramakrishna and K. Divakara Rao, Pramana J. Phys. 54, 255 (2000)]

The CBS causes an increased probability of return of the wave to the origin. For large l^*, its effect on the wave transport is small. For small I*, however, the angular range over which CBS is effective becomes very large, and this causes a renormalization of the diffusion constant, which is in fact a negative quantum correction to the classical Drude diffusion coefficient. This correction is, however, dependent on the sample size: because for a small sample, the CBS from very long paths would not contribute as they escape from the sample. The correction is proportional to $(1/L - 1/l^*)$, $\ln(l^*/L)$ and $l^* - L$ in one, two and three dimensions respectively [31, 7]. This negative correction to the conductivity (diffusion coefficient can make the conductivity go to zero for small enough I* and large enough size L, beyond which the diffusion picture is no longer valid. What happens then is a localization of the wave caused by disorder and multiple scattering - The Anderson localization [4]. The CBS is thus thought to be a precurser to the Anderson localization transition and is also known as weak localization.

We note that the time-reversal symmetry is a necessary condition for CBS. If the time reversal symmetry were broken, say, by applying a magnetic field for electrons

or by optical activity of the medium for light, then the enhancement factor reduces from 2 [32] and CBS becomes less effective for localization.

1.4.2 Anderson localization

The Anderson localization theorem states that for a non-interacting quantum system (one body problem) in a disordered medium, if the disorder strength is greater than a critical quantity, then all the eigen-states of the system are localized. This result was derived by Anderson by considering a tight-binding Hamiltonian on a 3-D lattice and disordered site energy [4]. The critical condition for the localization transition involves the strength of the disorder, the hopping strength and the co-ordination number – all the states are localized for $\Delta/W > (\Delta/W)_{critical}$, where A is the range of the disordered site energy and W $\equiv$ bandwidth $= 2ZV$, where V is the hopping strength and Z the coordination number. The result is very general and not restricted to this model alone. Localization means that if an electron is injected at a given point in the medium, then even after a long time $(t \to \infty)$, the probability of finding the electron decays exponentially away from the point of injection with a length scale known as the localization length (ξ_{loc}), i.e., $(\psi \sim \exp(-|\vec{r} - \vec{r_0}|/\xi_{loc})$. In this case, there are no propagating modes and no transport takes place, making the sample an insulator.

A simplistic way of thinking about the Anderson localization is the following dynamical argument. Consider an electron trapped at one of the lattice sites. This electron can hop on to the neighbouring sites and so on thus carrying a current. In order to hop the electron has to first find another suitable potential well with an energy level close to its original site energy. After all energy has to be conserved even though in the process of tunneling it can borrow energy temporarily from the "Heisenberg bank". As the disorder level increases, the probability of finding a suitable site nearby diminishes. While the number of distant neighbours increases algebraically with distance, the probability of tunneling to a far-off site, of course, decreases exponentially with distance. These considerations lead to a critical disorder beyond which no transport can occur. In this sense, the localization problem is similiar to the problem of classical percolation. It should, however, be stressed that the underlying cause of localization is wave interference and multiple scattering. For the localization to

occur, the scattering should be so strong that the wavelength of the wave should become comparable to the transport mean free path. This condition is known as the Mott-Ioffe-Regel (MIR) condition[33] and implies that multiple scattering becomes effective within the passage of a wavelength and the description in terms of propagation is no longer valid. With regard to the CBS, the CBS cone width now encloses almost all the backward direction $(\delta\theta \sim (kl^*)^{-1})$ and the probability of return to the origin is very high.

An important quantity in the theory of localization is the Thouless conductance [34]. Consider the eigenenergy levels of a d-dimensional hypercube of linear size L^d. If $\Delta E(L)$ be the average energy level spacing near the Fermi-level, and $\delta E(L)$ be the width of the energy levels caused by the hybridization with the free space continuum modes outside, then the conductance $G(L)$ of the hypercube measured in units of the quantum of conductance $e^2/\pi\hbar$ is the Thouless conductance $[g(L) \equiv G(L)/(e^2/\pi\hbar)]$ given by $g(L) = \delta E(L)/\Delta E(L)$. This is the analogue of the finesse factor defined in optics for a resonator cavity. When we put several such blocks together, then if $g(L) \ll 1$ at the scale of $L \sim l^*$ (the transport mean free path), the electron in one block will not penetrate another and we have an insulator (localization). On the other hand, $g(l^*) \gg 1$, there will be no barrier to transport and the system will be a conductor. In an influential paper, Abrahams et al. [35] assumed that $g(L)$ is the only scaling parameter that affects the behaviour as the blocks are put together, *i.e.*, as $L \to \infty$. This is the single parameter scaling assumption which states that the quantity which controls the scaling of the conductance with the system size is the conductance itself. Using this assumption alone, it was argued out that in the limit of large system size, there exist no extended states in two dimensions and a *fortiori* in one dimension. There is a continuous metal-insulator transition in three dimensions, with extended and localized states separated in energy by the Mobility Edge for the case of a sub-critical disorder, as indeed observed experimentally. This idea of the Mobility Edge for a subcritical disorder was first proposed by Mott [36], who, however, argued for a discontinuous metal-insulator transition implying a minimum metallic conductivity.

The result that all states are localized, for arbitrarily weak disorder, in a one-dimensional random medium is exact. This is simply a manifestation of the fact that

transport in 1-D can be described by transfer matrices that multiply in series and the product of random transfer matrices can be shown to yield exponential localization [37, 38]. A similiar results holds for the 2-D case too.

1.4.3 Localization of light

Although Anderson first proposed electron localization in 1958, the idea that classical waves such as light, acoustic waves or phonons could be localized as well, gained momentum only in the 1980's [6, 39, 40]. Light localization is attractive as it forms a non-interacting system in a medium with small optical non-linearity. Due to this reason it can offer a direct experimental verification of the scaling theory for pure disorder without interparticle interaction. Further, the bosonic nature of light allows for coherent absorption and amplification and makes it interesting to study. We will discuss this aspect in the next Section.

There are some important differences between light localization and electron localization. The wave equation governing the propagation of light (in a region without free charges)

$$\nabla^2 \vec{E}(\vec{r}) + (\epsilon_0 + \epsilon_{fluct})\frac{\omega^2}{c^2}\vec{E}(\vec{r}) = 0, \tag{1.11}$$

is similiar to the Schrödinger equation where $\epsilon_0\frac{\omega^2}{c^2}$ (ϵ_0 - the average part of the dielectric constant) plays the role of the energy eigenvalue and is always positive. The role of the spatially fluctuating random potential is taken by $\epsilon_{fluct}\frac{\omega^2}{c^2}$ (ϵ_{fluct} is the spatially fluctuating part of the dielectric constant). The dielectric constant is assumed positive implying that the energy of the electron in the equivalent case is positive. Thus, light localization corresponds to the equivalent case of localization of electrons with energy higher than the highest potential barrier.

Now, the scattering strength at low frequencies(long wavelength) becomes very small due to the w^2 factor multiplying the fluctuating potential (this corresponds to the λ^{-4} Rayleigh limit). The high frequency (short wavelength) limit, on the other hand, corresponds to geometrical or ray optics. Clearly, there cannot be localization, neither in the low energy Rayleigh limit due to weak scattering nor in the high energy geometrical optical limit where again the MIR condition cannot be satisfied, and we have extended states. Thus, an intermediate frequency window, between these two limits has been suggested for the observation of light localization. For example, use

of the enhanced Mie scattering cross-sections at the Mie resonances [41] and the plasmon resonances of metallic microparticles [42] have been suggested [2]. Sajeev john has suggested the localization of Bloch waves propagating in a photonic band-gap medium at the edge of a band-gap[43], where the Bragg scattering favours the MIR condition. In lower dimensions $D \leq 2$, however, all states will be localized in an infinite medium, for arbitrarily small disorder.

Experimentally, observation of the strong Anderson localization of light has been reported recently[44], in a densely packed highly scattering powder of GaAs semiconductor (sub-micron) particles. The transmission decayed exponentially with increase in thickness of the sample suggesting localization of light. Although, the authors claim to have taken precautions to minimize absorption in their samples which would also give an exponential decay of the transmittance, their claim has been strongly contested [45]. It is further unclear if the above behaviour is a purely disorder induced localization. A densely packed monodisperse powder would tend to get periodically stacked (whether a random close packing of spheres is possible, itself remains an open question [46]). Hence it is difficult to rule out the possibility of a Bragg stop band having played a role in the experimental observations.

1.5   Active random media

Now, we will turn to the case of active random media. By active media, we will mean in this thesis, coherently absorbing or amplifying media. Coherent absorption or amplification for a wave propagating in a medium is usually modelled by adding an imaginary part of the proper sign to the potential or the refractive index [47]. This is the case of coherent absorption or amplification, where the absorption/amplification occurs probabilistically and does not affect the phase coherence of the wave as an inelastic scattering process in general would. For the case of light, coherent amplification can occur due to the stimulated emission. Moreover, the coherent state of a laser is an eigenstate of the annihilation operator – removal of photons by absorption simply multiplies the state by a complex number, and hence the coherence is

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[2]However, the situation is more complicated here. The large cross-sections at these resonances affects the wave over considerably larger regions than the scatterer sizes, and for a dense system of scatterers leads to correlated scattering which can effectively reduce the scattering. Also it appears that the main effect of these scattering resonances is a renormalization of the group velocity of the propagating wave, caused by the large delay times of scattering near these resonances [160].

maintained. These are Bosonic properties without a Fermionic analogue. In the case of electrons, coherent amplification is not applicable. However, coherent absorption is possible in the sense qualified below. Of course, electrons cannot disappear in ordinary processes as the number of electrons are conserved. But, one can agree to consider only electrons in one coherent mode (channel). Inelastic scattering processes such as electron-phonon scattering will deplete the probability amplitude of electrons in this channel and that will appear as coherent absorption when we measure only the coherent part of the wavefunction by an interference measurement. This can also be modelled by adding extra side, or 'fake' channels for scattering through which the electrons can be lost from the system into electron reservoirs [48, 49]. This has been called *stochastic absorption*. This is in contrast to the *deterministic absorption* by a randomly operated chopper which blocks (absorbs) with complete certainty when the vanes intercept the beam. The coherence of the wave is not preserved in this case to the same extent [50].

We now ask the question, what happens if a disordered medium is made coherently absorbing/amplifying as well ? Would it destroy Anderson localization? Most studies indicate that it does not! In fact, localization in an absorbing medium has been shown to cause enhanced absorption/amplification [39, 55, 58] and the localization seems to get strengthened in amplifying media due to enhanced CBS – involving prolonged return path lengths [51, 87]. This also brings up the recent phenomenon of lasing in such random amplifying media (RAM), which we will discuss here.

1.5.1 Random lasers

First of all, let us consider what happens in a conventional laser. The amplifying (homogeneous) medium is placed in a one-dimensional optical cavity, which traps the light emitted spontaneously by the excited medium, forcing it to repeatedly pass through the amplifying medium while undergoing amplification due to stimulated emission in each traverse. Laser action occurs when the amplification in each round trip exceeds the loss at the cavity output couplers and the optical elements in the cavity. Then a coherent mode builds up in the cavity and results in a unidirectional, monochromatic, coherent beam. The frequency of the laser light is at an eigen-frequency of the cavity, near the maximum of the gain spectrum of the amplifying

medium. Thus, the laser action occurs due to a resonant coherent feedback of light from the mirrors. We note that even the presence of a small inhomogeneity, or disorder in the cavity can cause mode-hopping, intensity fluctuations etc. and can be a major source of noise in an otherwise highly ordered output. Can this seemingly antithetical nature of disorder and lasing be reconciled and can lasing be actually aided by the deliberate introduction of scatterers into a cavity? This question has evoked much scientific debate for the greater part of the past decade.

It all began with the experimental discovery [16] of the original proposition by Letokhov[52] in 1968, that placing random scatterers in a gain medium could enhance the frequency stability of the laser emission. In these experiments, it was found that the introduction of disorder (by suspending titania microspheres) into a homogeneous Rhodamine laser dye solution caused a drastic spectral narrowing of the emission from the dye above a well-defined threshold of pump light intensity. Sure, this is a well-known phenomenon which occurs even in homogeneous amplifying media due to amplified spontaneous emission (ASE). But the remarkable aspect of these experiments was that, the threshold of the pump laser intensity at which the flourescence spectrum collapsed dramatically was almost two orders of magnitude smaller in the case of the microsphere-laser dye suspension, compared to the case of the ASE in the neat laser dye solution. A typical experiment seems deceptively simple. Take a beautiful laser crystal and grind it into a fine powder (size $\sim 1\mu m$). Now excite this powder by pumping it in the absorption band with pulses from a powerful laser. Spectral and temporal measurements of the emission from the powder in different directions are carried out. Another option would be to suspend the scattering microparticles such as titania or polystyrene microspheres in a laser dye solution, or to imbed the scatterers into a laser dye-doped polymeric matrix.

Now, let us look at a multiply scattering amplifying medium with weak, dense scatterers, where the diffusion picture holds. The light emitted spontaneously by the medium will have to undergo a long random walk inside the medium (which could be orders of magnitude longer than in a homogeneous medium) before exiting. During every part of this random walk, the light is continuously being amplified due to stimulated emission. Thus, gain can become larger than loss due to leakage at the system boundaries and one can have lasing action caused by the diffusive feedback.

The output loss scales as the surface area of the medium through which the light exits while the gain scales as the overall volume of the medium. As the size of the RAM is increased beyond a point, the gain becomes larger than the loss and a strong flash of light will occur in all directions. This is exactly analogous to what happens in a fission nuclear bomb or reactor where neutrons are scattered and amplified by nuclear fission (multiplied by chain reactions) – the system becomes supercritical beyond a critical size of the system. In such random laser, one notes that there will be a large gain narrowing of the emitted spectrum as the photons at the emission maximum would be much more amplified in their long passage through the medium. Thus, it is a non-resonant "feed-forward mechanism" here that causes lasing action compared to the feedback mechanism in a conventional laser. It is this large extension of the path length in the RAM due to multiple scattering that is responsible for the lowering of the threshold pump intensity. Note that in comparision to the unidirectional output of a conventional laser, the random laser will emit in all directions. In Fig.1.6, experimental spectra obtained from a RAM (consisting of polystyrene microspheres suspended in a Rhodamine 6G solution) obtained below (curve-a) and above (curve-b) the threshold of lasing are shown. Above the threshold, the FWHM of the spectrum is about 6nm. Also, due to the very large intensities which occur during the lasing, the population inversion in the RAM is quickly depleted (or 'bleached'), leading to a sharp temporal peak or pulse shortening of the emission. If the exciting pump beam is still present, then the population inversion will again build up and deplete successively, resulting in a train of spikes in the emission.

Coming to the other details of the emission, it was found that at some intermediate pump powers, a bichromatic emission results (See Fig.1.6, curve-c). This has been explained as a result of the displaced absorption and emission spectra of the RAM, based on Monte Carlo simulations [53]. Another extremely interesting feature observed in these media is that there is a sharp reduction in the threshold of pumping even when the transport mean free path of light is much larger than the sample size[54]. This would naively imply that there are no multiple passes through the medium as the photons escape ballistically. This effect could, however, be explained [54] by considering the effect of the probabilistically rare sub-mean-free-path large-angle scattering events, which are now rendered important by the virtue of high gain
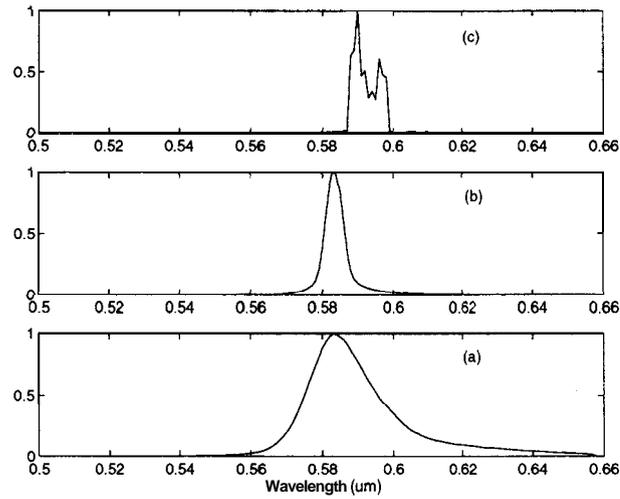
Figure 1.6: The normalized spectral emission from a RAM consisting of polystyrene microspheres suspended in a Rhodamine 6G solution (a) sub-threshold (b) above-threshold (c) bichromatic emission at intermediate pump levels. The experimental data is the due to the courtesy of S. Mujumdar [53].

in the medium. Thus, a manifestation of the well known St. Petersburg paradox, wherein rare events but with a very large multiplier value can become more important than the more frequent events but with a small multiplier value.

Finally, we look at very strongly scattering ($kl^* \sim 1$) and amplifying media where the diffusion picture breaks down and localization effects become important. Also, in the above picture of light diffusion with gain, the amplification could very well have been an incoherent process (such as in the generation of neutrons in a nuclear fission reaction where the outgoing neutrons have no phase correlations with the incoming neutrons). Stimulated emission is, however, a coherent process where the emitted photons have the same phase and direction as the incoming photon. In fact, this is a bosonic "stimulation" property without a fermionic analogue. This property of coherent amplification of light coupled with the coherent feedback offered by the Anderson localization can result in a new kind of coherent laser output. This enhancement of amplification due to a synergy between coherent amplification and wave confinement by localization was first theoretically predicted by Pradhan and Kumar [55] in 1994 and subsequently confirmed by several others[56, 57, 58, 59]. In this case, the recurrent multiple scattering events forming closed loops[91] can provide feedback much like in the ring cavity lasers. Of course, due to the random nature of the

medium, there would exist a distribution of many such random cavities with different Q-factors. As the pump laser intensity is increased, the laser threshold condition (gain = loss) would be satisfied first in the cavities with the highest Q-factors. Laser oscillation would occur at frequencies determined by the cavity resonances. Thus, the laser emission would consist of sharp discrete lines. At higher pump intensities, oscillations would begin even in lossier cavities and more lines would be added to the emission spectrum. One notes that the random cavities formed by the recurrent multiple scattering would be totally different in different samples Thus, the lasing lines would vary from sample to sample, a typical effect found in disordered systems. Also, the random cavities would have outputs in different directions and the emitted spectrum would be different in different directions and we have a multi-mode laser emission in all directions. In the recent experiments of Cao et *al.*[60, 61] with very strongly scattering semiconductor powders (Zno, GaN), all the above effect were observed to occur. The lasing lines had extremely small spectral linewidths(~ 0.2nm). Thus, a random laser with coherent feedback was claimed. One should note that, in these experiments the semiconductor powders were deposited on glass substrates as a thin film of powder ($\sim 10 \mu m$). Hence, keeping in mind the large dielectric constant of the semiconductor material, it is possible that total internal reflection at the interfaces could have effectively acted as mode-guiding mechanism, thereby effectively reducing the dimensionality of the random system to two. Thus, there are doubts that the fine spectrum observed in these experiments could be due to a reduced dimensionality of the system when the localization effects are easier to achieve. In their recent work on spatial confinement of light [61], Cao et al. show that the lasing volumes can be as small as $2 \mu m$, almost comparable to the wavelength. This seems to suggest that morphological resonances could be the reason.

We note two additional points. The random laser with the coherent feedback provided by Anderson localization should be distinguished from the traditional distributed feed-back laser [62]. In the latter case, the feedback is derived from a periodic modulation of the gain/refractive index, the period of which satisfies a condition of resonance at the laser wavelength. By comparision, the feedback due to Anderson localization is coherent, but non-resonant. The second point is that, in a one-dimensional RAM, the system never really becomes a laser (oscillator) and remains

only an amplifier. This is because the localization length sets an effective cavity length over which the coherent output develops. The threshold condition is that the amplification length (l_{amp}) should become larger than the localization length (ξ_{loc}). This condition is never really satisfied because the amplification enhances localization and the effective localization length ($l_{eff}^{-1} = \xi_{loc}^{-1} + l_{amp}^{-1}$) reduces with increasing amplification [57, 63].

1.6 Sojourn time in a scattering potential

Till now we have been discussing the static aspects of scattering. Now we turn to the dynamic aspect of scattering, namely, the time delay undergone by a wave in the process of being scattered by a potential. Obviously, this time is related to the actual time of sojourn in the scattering region. For the case of a classical particle, both the times will be one and the same. But it is not obvious that the equality would hold for a wave. This time of sojourn (also called the dwell time) is of interest for mesoscopic systems.

This issue of the time of sojourn raises some fundamental questions regarding role of time in quantum mechanics. There are two distinct question: (i) where is the particle at a given time 't'?, and (ii) at what time (if any) is the particle in a given region of space? The former corresponds to a position measurement, for which we have a well-defined position operator. The latter is the arrival time for which it has not been possible to define a self-adjoint operator, in general [64] [3]. Thus, the quantity in question does not appear to be an observable. The arrival time is, of course, not the parametric time that appears in the Schrodinger equation. It (the arrival time) is a dynamically determined quantity, determined by the conditions of scattering in the region of interest. This quantity appears to be measurable as it can be determined experimentally. But it seems dependent on the nature of the experiment. Thus, there is no accepted single definition of this quantity and this has resulted in several

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[3]It is possible to define an operator by first considering the classical time taken by a particle to propagate from one point (x_0, p_0) in the phase space to another (x, p), and then quantizing the classical dynamical variables. But the procedure would not work for barrier tunneling, where there is no classical path linking the initial and final points. In that case, we would have to define an imaginary velocity which would not allow the 'time operator' to be Hermitian. In another approach, it is often stated that one can define a Hermitian operator Θ, which measures whether a particle is in a given region of space or not [65, 66]. But a procedure of practically implementing the operator is yet to be found.

timescales defined in different manners. First of all, different time-scales can be defined depending on the input and output channels of scattering. These are the *conditional times* and in the 1D case, we have the reflection and transmission sojourn times. In the following, we will briefly review a few of the important timescales. For recent reviews of the topic, we refer the reader to Refs.[66, 67, 68].

One of the most common approach is to follow any fiducial feature or marker such as the peak of a wavepacket as it scatters off a potential. This view was first taken by Wigner[69] who defined the delay time in terms of the energy derivative of the phase of the scattered wave $\tau_w = \hbar(\partial\theta/\partial E)$. This procedure is, however, rendered meaningless when the scattering potential strongly distorts the wave packet. In particular, Büttiker and Landauer [70] have stressed that there is no causal connection in the incoming peak transforming into the peak of the outgoing wave. The same applies to the centre of mass of the wavepacket. Another wavepacket based approach, proposed by Stevens, is to follow the forward edge of the wavepacket with a sharp cut-off [71]. But the problem of associating the particle's position with the deformable wave-front remains.

A second method of dealing with the sojourn time is to define it in terms of the probability of finding the particle inside the spatial region of interest. Thus, we have an average time of dwell (the Smith dwell time)[112] in the region of interest $[0, L]$, regardless of whether it is eventually transmitted/reflected:

$$\tau_d = \frac{1}{j} \int_0^L |\psi(x)|^2 dx, \tag{1.12}$$

where j is the incoming flux and $\psi(x)$ is the wave function in the stationary scattering picture, *sub specie aeternetatis.* Also for a time-dependent case (pulse), the average dwell time is defined as

$$\tau_d = \int_{-\infty}^{\infty} dt \int_0^L |\psi(x, t)|^2 dx, \tag{1.13}$$

Closely related to this, is the path integral approach of Sokolovski and Baskin [72], to describe the time of dwell in terms of a Feynman path integral over every path linking the incidence and the subsequent transmission/reflection of the particle. Yet another approach is to use the WKB wavefunction in the barrier region [73], where one can identify a forward and a backward velocity field from the wave function and hence, define an appropriate traversal time. This approach has been used in Section **3.4.**

1.6.1 Quantum clocks

A third option is to look beyond these phase delay and dwell time definitions based on the wave function and consider meaningfully the dynamical evolution of an extra degree of freedom ('clock') attached to the particle due to local interaction with a potential applied in the region of interest. The effect of the interaction on the clock would depend on the time of sojourn in the region of interest. The external potential, or field, is made as small as possible to avoid any effects on the propagation of the particle in question. Three such clocks have been proposed and will be reviewed briefly here.

(i) The Büttiker-Landauer oscillating barrier

Here the original static potential is augmented by a small oscillation in the potential height or width. At low frequencies of the oscillation, the particle sees essentially a static potential with the scattering amplitudes (transmission/reflection) slowly varying in time (the adiabatic regime). At high frequencies of oscillations, the particle no longer sees a static potential, but is affected by many cycles of the modulation during its interaction with the potential. The particle begins to absorb/emit quanta and the scattered wave develops energy sidebands (the diabatic regime). The point of crossover of this adiabatic-diabatic behaviour yields a timescale which is taken to be the interaction time of the particle with the potential. For an opaque symmetric rectangular barrier, the barrier traversal time turns out very reasonably to be $\tau_{BL} = mL/\hbar\kappa$, where L is the barrier width, and $\hbar\kappa$ is the imaginary momentum in the barrier region. This τ_{BL} will henceforth referred to as the Buttiker-Landauer time in this thesis.

(ii) The Larmor clock

Buttiker reconsidered the idea of using the accumulated spin precession angle of a spin in a magnetic field [75], earlier proposed by Baz' and Rybachenko[76], as a clock for the tunneling time. The proposed experimental situation, where a spin initially polarized along the x-axis moves along the y-axis through the potential region in which a magnetic field along the z-axis is present, is schematically depicted in Fig. 1.7. Büttiker recognized that there is a tendency of the spin to align along the magnetic field (spin rotation) in addition to the Larmor precession in the plane perpendicular to the magnetic field (spin precession). Biittiker associated time scales in relation to
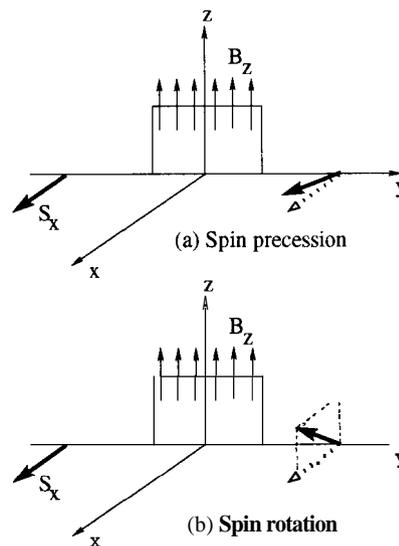
Figure 1.7: A schematic depiction of spin rotation and spin precession in the Larmor clock

the extents of spin rotation(τ_z) (which is, in fact, dominant for an opaque barrier) and spin precession (τ_y), and defined the traversal(reflection) time to be the Pythogorean sum $\sqrt{\tau_y^2 + \tau_z^2}$. The physical basis of associating these quantities to the traversal time is not clear and has been questioned [77, 68]. For an opaque rectangular barrier, the Larmor time for transmission becomes the same as the Biittiker-Landauer time.

(iii) Absorption as a clock

The use of absorption as a clock was originally proposed by Pippard in a private communication to Büttiker[78], who subsequently examined it [78], along with Golub *et* al. [79] and Wang *et al.*[80]. The amount of absorption suffered by a wave in the presence of an imaginary potential was associated with the time dwelt in the given region, where the imaginary potential was present. It turned out that for a symmetric rectangular barrier this time was identical to the average Smith dwell time and consequently absorption was thought to be incapable of distinguishing between reflection and transmission [78, 77].

Most of these approaches have associated problems, such as they yield complex times, or negative times for certain deterministic potential configurations. Perhaps, the clocks affect the spatial motion itself [79, 77]. the Feynman path integral approach yields a complex time, though the real and imaginary parts of the time appear to be related to the Larmor times. The phase delay, of course, can go negative as it

only compares the phase shift in the presence of a potential to the case with no potential. However, the other times given by the clocks, defined more as interaction times, should remain positive. But that is not the case. So it begs the question if it is meaningful at all, to talk about these times and is it necessary to bother about these times? When we think of the time of sojourn of a particle in a given region of space, we are merely taking an alternative viewpoint in quantum mechanics. Obviously the sojourn time cannot provide answers to questions which cannot be answered within quantum mechanics. Almost all questions in the realm of quantum mechanics can be discussed without recourse to the concept of the sojourn time. However, the sojourn time is a useful practical and conceptual tool, given our classical intuition about time. It is all a matter of wanting to compare timescales. But in order to do so, we need a *prescription* for the estimation of the sojourn time, which yields a *reasonable answer* to be useful as a *practical and conceptual tool.* for this to happen, in our opinion, the sojourn time should be: (i) real, (ii) positive, (iii) additive (times so calculated for different parts should add up to give the total time), (iv) calculable, and (v) measurable, even if not observable as an operator in quantum mechanics.

We have hopefully provided such a prescription in Chapter-3 of the thesis. In this context, we should perhaps mention the stochastic quantization of Nelson [81], where there are real stochastic paths associated with quantum motion and the average of the time spent by a particle traversing these paths can be taken to be the sojourn time. Obviously, this time will stay real and positive, and this view has recently been explored[82]. Our prescription, by comparision, is within the conventional framwork of quantum-mechanics.

1.7 Fluctuations and statistics

As described earlier, there are several quantities in mesoscopic physics which fluctuate, often violently, as a function of the system parameters such as the length, potential strength, the presence of a magnetic field and so on. Examples are the resistance fluctuations in a mesoscopic conductor as a function of an applied magnetic field[83], non-periodic spatial fluctuations within a speckle pattern and the spectral fluctuations within a single speckle spot observed in microwave and optical scattering. These fluctuations are reproducible (not stochastic). This fluctuating behaviour is a

universal aspect of wave propagation in random media. Needless to say, it is wave interference that is again at the heart of these fluctuations. In this context, there two kinds of statistical quantities:

(i) Self averaging quantity

Many thermodynamic quantities are of this kind. A statistically fluctuating quantity is called self-averaging, if the average of the quantity and the r.m.s fluctuations in the quantity grow with the system size (or number of particles), such that in the (thermodynamic) limit of large system size, the ratio $\frac{r.m.s\ fluctuation}{average} \rightarrow 0$. Examples are the pressure and temperature of a classical gas, density of states, classical resistance etc. In this case, the central limit theorem would apply and we can talk sensibly about the system and the statistical quantity using the mean value and fluctuations about the mean.

(ii) Non-self-averaging quantity

In contrast, the r.m.s fluctuations of some quantities increases with the system size faster than its average value, so that the ratio $\frac{r.m.s\ fluctuation}{average} \not\rightarrow 0$, or can increase without bound in the limit of a large sample size. The quantity of interest now depends on the detailed inner configuration of the sample. The quantum conductance of a mesoscopic sample is a canonical example where the conductance is different for sample with differing impurity configurations and same impurity (average) concentration. Indeed, here we have a Universal Conductance Fluctuations (UCF) $\sim C_d(e^2/\pi\hbar)$, where C_d depends only on the dimensionality [84]. Non-self-averaging behaviour is a manifestation of wave interference. When a quantity displays non-self-averaging behaviour, we cannot meaningfully characterize the quantity by the mean and the fluctuations about the mean, but would require the entire statistical distribution of the quantity. Examples are the conductance/resistance of a quantum resistor, reflection/transmission coefficients and the delay time of scattering in a disordered medium etc. One often resorts to considering the distribution of the logarithm of such fluctuating quantities as the latter often obeys a central limit theorem because of the levelling property of the logarithm [155]. The log-normal conductance fluctuations in a 1D disordered wire is a good example of this behaviour[85].