Sources and gain in photonic random media
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Citation for published version (APA):
El-Dardiry, R. G. S. (2012). Sources and gain in photonic random media

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CHAPTER 1

Introduction

The central topic of this thesis concerns the interaction between sources of electromagnetic radiation and complex multiple scattering media. Both of these two elements span a vast field in the natural sciences themselves. Our treatment of the matter in this introductory chapter is aimed at putting forward only the essential physics and background required to understand the experiments and theory described in this thesis. First, the various mechanisms for generation of electromagnetic radiation are described in Sec. 1.1. Second, the physics behind both single and multiple scattering of waves is briefly reviewed in Sec. 1.2. In Sec. 1.3, random lasers and infinite range correlations are introduced as two central research subjects that combine sources and multiple scattering. The contents of this thesis and our contribution are then outlined in Sec. 1.4.

1.1 Sources of electromagnetic radiation

Due to its importance in the everyday human perception of the world, light has inevitably been studied and put to use since ancient times. The wave nature of light was put forward by several natural philosophers of whom Christiaan Huygens (1629-1695) is generally accredited the most. His *Traité de la lumière* provides a fascinating look onto the status of optics during the early stages of the modern era [1]. By drawing an analogy with sound waves, Huygens concludes that light must be a wave that propagates via the mediation of tiny masses. In the original formulation of the famous principle named after him these tiny masses, the ether, indeed feature prominently:

*De sorte qu’il faut qu’autour de chaque particule il se fasse une onde dont cette particule soit le centre [1] (So it arises that around each particle there is made a wave of which that particle is the centre [2]).*

This formulation has often been rephrased in terms of secondary sources instead of particles, although it would technically be more correct to view the “particles” as scatterers
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since no new light is generated. In fact, Huygens postulated that the generation of light was the consequence of matter in rapid motion which, as we shall see shortly, is not far from our modern day understanding of light sources. The intuitive concept of an ether remained prevalent among scientists, but had finally to be refuted due to experimental results obtained by Michelson and Morley at the end of the nineteenth century. Their experiments indicated that the speed of light is constant thereby contradicting predicted anisotropies from ether based theories.

However, the wave concept of light has remained as one of the foundations of modern science and technology. The work of Faraday and Maxwell led to the discovery that light is an electromagnetic wave whose characteristics are well described by solving the Maxwell equations. These four equations describe how the electric field $E$ and the magnetic field $H$ depend on the volume density of electric charge $\rho_c$, the polarization of a medium $P$, the magnetization of a medium $M$, and the current density $J$ [3]:

$$\nabla \cdot E = -\frac{1}{\epsilon_0} \nabla \cdot P + \frac{\rho_c}{\epsilon_0}, \quad (1.1)$$
$$\nabla \cdot H = -\nabla \cdot M, \quad (1.2)$$
$$\nabla \times E = -\mu_0 \frac{\partial H}{\partial t} - \mu_0 \frac{\partial M}{\partial t}, \quad (1.3)$$
$$\nabla \times H = J + \epsilon_0 \frac{\partial E}{\partial t} + \frac{\partial P}{\partial t}. \quad (1.4)$$

Here $\epsilon_0$ and $\mu_0$ are the permittivity and permeability of the vacuum. In this thesis only nonmagnetic electrically neutral media are considered, therefore $M = 0$, $\rho_c = 0$, and $B = \mu_0 H$. By taking the curl of Eq. (1.3) and then inserting Eq. (1.4) on the right hand side, the equation for electromagnetic waves is found

$$\nabla \times (\nabla \times E) + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P}{\partial t^2} - \mu_0 \frac{\partial J}{\partial t}, \quad (1.5)$$
$$-\nabla^2 E + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P}{\partial t^2} - \mu_0 \frac{\partial J}{\partial t}. \quad (1.6)$$

Where in the last step the vector identity $\nabla \times (\nabla \times V) \equiv \nabla (\nabla \cdot V) - \nabla \cdot (\nabla V)$ was used together with $\nabla \cdot E = 0$, and $c = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light in vacuum. Wave Eq. (1.6) lies at the heart of photonics research. It is used, e.g., in the design of antennas on the nanometer scale [4–6], the development of higher resolution microscopes [7], the discovery and fabrication of optical metamaterials [8–10], and the understanding of the propagation of waves in complex photonic media [11]. With increasing complexity of photonic structures, solving the equation numerically can be a tremendous computational exercise and we shall often find the need to simplify matters considerably.

The right hand side term in Eq. (1.6) shows that electromagnetic waves are scattered, generated or absorbed by the acceleration of charges. The acceleration of charges can either take place by a changing current, $\mu_0 \frac{\partial J}{\partial t}$, as is the case in metals, or a change in the polarization of the medium, $\mu_0 \frac{\partial^2 P}{\partial \tau^2}$, as is the case in insulators, or by a combination of both as is the case in semiconductors. The frequency of the wave determines the type of electromagnetic radiation that is generated. Light forms just a small portion of the entire electromagnetic spectrum. Although light has the obvious advantage of being visible, it requires photonic structures to be in the nanometer range making the construction of these structures a serious technological challenge. Waves with a smaller frequency than light
1.1. Sources of electromagnetic radiation

enable us to study similar phenomena on larger length scales while the underlying physics remains the same. In this thesis we describe experiments performed with visible light (400-790 THz) and microwaves (10 GHz).

To first order, electromagnetic sources are described by oscillating dipoles. An isotropically radiating point source is well approximated in the far field by an oscillating dipole. A classical dipole is therefore one of the most frequently used sources in photonics. In Sec. 1.1.1, the expressions of the fields for a point dipole are derived following a standard reference [12]. Oscillating dipoles can be induced in a variety of ways depending on the desired frequency. Dipole sources can be created by inducing oscillating currents in an antenna in the case of microwaves or by thermal and spontaneous emission in the case of light. The latter will be discussed in Sec. 1.1.2 and Sec. 1.1.3, and will be followed by a discussion on stimulated emission and absorption in Sec. 1.1.4.

1.1.1 Radiating point dipole

The wave equation deduced from the Maxwell equations showed that sources of radiation stem from either accelerating charges or changing currents. The oscillating dipole is the easiest example of such a source and it is frequently used to describe experimental configurations with atomic and molecular light sources. In this section the expressions for the fields of a radiating point dipole are derived [12] at a distant point \( r \) away from the dipole.

Consider two metal spheres connected by a thin wire of length \( d \) aligned along the \( z \)-axis with its center located at the origin as illustrated in Fig. 1.1(a); one sphere contains a charge \( q(t) \), while the other sphere contains an opposite charge \(-q(t)\). The charge on the spheres oscillates with angular frequency \( \omega \). The two spheres then form a dipole with oscillating dipole moment \( \mathbf{p}(t) = p_0 \cos(\omega t) \hat{z} \) where the amplitude is given by \( p_0 = q_0 d \). For an ideal point dipole it is assumed \( d \ll \lambda \ll r \). In electrodynamics, the electric and magnetic fields at point \( r \) can be expressed in terms of the potential \( V \) and the vector potential \( \mathbf{A} \)

\[
\mathbf{E} = -\nabla V - \frac{\partial \mathbf{A}}{\partial t},
\]

\[
\mathbf{B} = \nabla \times \mathbf{A}.
\]
The retarded potential of the dipole is given by

\[ V(\mathbf{r}, t) = \frac{1}{4\pi\varepsilon_0} \left\{ \frac{q_0 \cos \left[ \omega \left( t - r_+/c \right) \right]}{r_+} - \frac{q_0 \cos \left[ \omega \left( t - r_-/c \right) \right]}{r_-} \right\}, \tag{1.9} \]

\[ V(\mathbf{r}, \theta, t) \approx -\frac{p_0 \omega}{4\pi\varepsilon_0 c} \left( \frac{\cos \theta}{r} \right) \sin[\omega(t - r/c)]. \tag{1.10} \]

The second equation is the result of applying geometrical and Taylor approximations to Eq. (1.9) that follow from the criterion \( d \ll \lambda \ll r \). The vector potential is given by the integral of the current density over the volume of the wire.

\[ \mathbf{A}(\mathbf{r}, t) = \frac{\mu_0}{4\pi} \int \frac{\mathbf{J}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}', \tag{1.11} \]

\[ \mathbf{A}(r, \theta, t) \approx -\frac{\mu_0 p_0 \omega}{4\pi r} \sin[\omega(t - r/c)] \hat{\mathbf{z}}, \tag{1.12} \]

\[ = -\frac{\mu_0 p_0 \omega}{4\pi r} \sin[\omega(t - r/c)] (\cos \theta \hat{\mathbf{r}} - \sin \theta \hat{\theta}). \tag{1.13} \]

In the second step we used \( |\mathbf{r} - \mathbf{r}'| \approx r \) when \( d \ll r \). Putting the expressions for \( V \) and \( \mathbf{A} \) into Eqs. (1.7) and (1.8) and discarding all higher order terms in \( 1/r \), returns the radiated electric and magnetic field by an ideal dipole.

\[ \mathbf{E}(r, t) = -\frac{\mu_0 p_0 \omega^2}{4\pi} \left( \frac{\sin \theta}{r} \right) \cos[\omega(t - r/c)] \hat{\mathbf{\phi}}, \tag{1.14} \]

\[ \mathbf{B}(r, t) = -\frac{\mu_0 p_0 \omega^2}{4\pi c} \left( \frac{\sin \theta}{r} \right) \cos[\omega(t - r/c)] \hat{\mathbf{\phi}}. \tag{1.15} \]

From these two equations, the radiated intensity is found by calculating the cycle-averaged Poynting vector \( \mu_0 \mathbf{S} = (\mathbf{E} \times \mathbf{B}) \)

\[ \langle \mathbf{S}(r, \theta) \rangle = \left( \frac{\mu_0 p_0^2 \omega^4}{32\pi^2 c} \right) \frac{\sin^2 \theta}{r^2} \hat{\mathbf{r}}. \tag{1.16} \]

From this equation one understands that the intensity profile is shaped like a donut and the intensity falls off with \( r^2 \). In order for the dipole to act as a true source of radiation \( \oint \langle \mathbf{S} \rangle d\mathbf{a} > 0 \) where the integration runs over a closed surface containing the dipole, in words this condition implies radiation is created by the source. The divergence theorem, \( \oint \langle \mathbf{S} \rangle d\mathbf{a} = \int \nabla \cdot \langle \mathbf{S} \rangle d\tau \), allows us to analyze the divergence of the Poynting vector instead. Since \( \nabla \cdot r^{-2} \hat{\mathbf{r}} = 4\pi \delta(\mathbf{r}) \) with \( \delta(\mathbf{r}) \) the Dirac delta function, the divergence of a classical dipole reads

\[ \nabla \cdot \langle \mathbf{S} \rangle = \frac{\mu_0 p_0^2 \omega^4}{12\pi c} \delta(\mathbf{r}), \tag{1.17} \]

from which we conclude a classical point dipole indeed acts as a source for electromagnetic radiation. However, reciprocity implies that a classical dipole can also be excited by radiation. An excited dipole will of course also radiate. Conservation of energy dictates that for an excited dipole without dissipation \( \int \nabla \cdot \langle \mathbf{S} \rangle d\tau = 0 \). Hence, the oscillating dipole, although described by the same equations, has turned from a source into a scatterer. Scattering objects that are much smaller than the wavelength are well described by dipole scattering and are referred to as Rayleigh scatterers [13]. The physics of these relatively small scatterers is all around us and can be witnessed every day. The frequency dependence of the scattered power in Eq. (1.16) leads to the blue color of the sky, while the angle dependence causes scattered light from the sky to be partially polarized. We will come back to the physics of scattering particles in Sec. 1.2.1.
1.1.2 Spontaneous emission and vacuum fluctuations

Since the frequency of dipole radiation is simply given by the frequency of the oscillating charges, low frequency waves can easily be generated electronically at arbitrary frequencies with the right circuit design. The engineering and fabrication of antennas has become a major industry that covers the low end of the electromagnetic spectrum consisting of radio waves and microwaves. The difficulty of fabricating higher frequency sources by this method is one of the main reasons for the fact that the THz part of the electromagnetic spectrum was relatively hard to study in the past [14]. For infrared and visible light, high frequency oscillating dipoles can be formed by oscillating electron clouds in molecules and atoms as illustrated in Fig. 1.1(c) [15]. In nature, the main source for accelerating charges that emit light is due to thermal excitation. Every object with a nonzero absolute temperature radiates electromagnetic waves in thermal equilibrium, the sun and incandescent light bulbs are two everyday examples of these so-called black body radiators. Spontaneous emission of radiation can occur when excited atoms and molecules fall back to a lower energy state.

Until now we have described the physics of electromagnetism classically. However, spontaneous emission requires a quantum mechanical treatment of both the field and the atom in order to be in correspondence with experiment [16, 17]. A semiclassical description of spontaneous emission, in which the fields are treated classically and the atom quantum mechanically, essentially fails because according to such a model an atom in the excited state $|\psi_2\rangle$ is in an unstable equilibrium and would therefore never fall back to the ground state $|\psi_1\rangle$ [16]. In quantum electrodynamics even vacuum modes contain fluctuating electromagnetic fields that perturb an excited atom. These perturbations will sooner or later force the atom to leave its unstable equilibrium and to undergo a transition to the ground state while simultaneously sending out radiation in the form of a photon\(^1\). The precise moment an atom decays by spontaneous emission is unpredictable, yet the average rate at which an atom decays is well described by quantum electrodynamics. Obviously the higher the number of modes that interact with the atom, the shorter it takes on average for an atom to decay. When the electromagnetic modes form a continuum, the average radiative decay of an excited atom at position $\mathbf{r}$ and transition dipole moment $\mathbf{d}$ is described by an exponential of which the decay rate $\gamma_r$ is given by Fermi’s Golden Rule

$$\gamma_r(\mathbf{r}, \omega, \hat{\mathbf{d}}) = \frac{\pi d^2 \omega}{\hbar \epsilon_0} \rho(\mathbf{r}, \omega, \hat{\mathbf{d}}). \tag{1.18}$$

In this expression, $\rho$ is the local density of electromagnetic radiative states (LDOS). The LDOS gives the number of states per unit volume to which an atomic oscillating dipole positioned at $\mathbf{r}$, oriented along $\hat{\mathbf{d}}$, and with frequency $\omega$ can couple.

Fermi’s Golden Rule yields another cornerstone of the field of photonics. The first part of Eq. (1.18) $\frac{\pi d^2 \omega}{\hbar \epsilon_0}$ is an atomic factor, which in practice can hardly be altered without operating at the Angstrom scale. However, by engineering the LDOS photonically one can gain control over the emission characteristics of an emitter with structures on the order of a wavelength [18]. The LDOS might appear an exotic quantity at first glance, yet a plain mirror can already change the LDOS significantly. The decay rate of an atom in proximity

\(^1\)Since spontaneous emission is a fully quantum mechanical process, the term photon in this context is justified. Occasionally and mainly to increase the readability of the text, we shall use the term photon when in principle a semiclassical description suffices and there is in principle no need for speaking about photons instead of energy density.
Figure 1.2: The emission spectrum from a collection of molecules or atoms can be broadened by (a) homogeneous broadening processes or (b) inhomogeneous broadening processes, or a combination of both. In the case of homogeneous broadening the linewidth is broadened for each individual emitter (gray lines) in a similar fashion. The output spectrum for a collection of emitters (black line) is therefore identical to the output spectrum of an individual emitter. Inhomogeneous broadening is caused by emitters having different center frequencies (gray lines). (c) In a four-level system the emitter quickly relaxes to the lower level of the excited state (wiggly arrow) from which it can undergo a radiative transition (straight arrows) to several vibrational sublevels of the ground state. From these vibrational sublevels the molecule relaxes back to the lowest level of the ground state. The spread in sublevels of the ground state is responsible for a significant homogenous broadening of the emission spectrum.

(∼λ) of a mirror oscillates as a function of its separation distance [19, 20]. In 3D photonic crystals with a high enough photonic contrast the LDOS vanishes completely leading to a photonic band gap in which atoms theoretically cease to decay at all. Recently it was shown experimentally that a photonic band gap can reduce the average decay rate by more than a factor 10 [21].

1.1.3 Non-radiative decay: quantum efficiency and spectral broadening

Besides undergoing a radiative transition from a higher energy state to a lower energy state via the process of spontaneous emission, atoms and molecules can, in general, also relax to a lower energy state by non-radiative transitions. Examples of processes that cause non-radiative transitions include lattice vibrations and molecular collisions [15, 16]. The quantum efficiency \( \phi \) is a quantitative measure for how much energy flows into the radiative decay channel in comparison to non-radiative decay channels and is defined by

\[
\phi \equiv \frac{\gamma_r}{\gamma_r + \gamma_{nr}},
\]

where \( \gamma_{nr} \) is the non-radiative decay rate. From an experimental point of view high quantum efficiency sources are often the emitters of choice in photonics. Not only because they simply return more light, but also because they are more convenient probes for measuring changes in the LDOS by performing lifetime measurements. A change in the LDOS results in a different value for the radiative decay rate and thereby changes the ratio between radiative and non-radiative decay. However, when the non-radiative decay channels are strongly dominating over the radiative channel, any change in the radiative decay rate will be hard to detect since lifetime measurements probe the total decay rate \( \gamma_{tot} = \gamma_r + \gamma_{nr} \), which is then hardly affected.

Non-radiative transitions within an emitter also have a significant impact on its emission spectrum. The spectrum of an ideal two-photon emitter with amplitude decay rate \( \gamma_{tot}/2 \) is given by the Fourier transform of an exponential which has a complex Lorentzian line shape.
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with a characteristic Full-Width at Half-Maximum (FWHM) linewidth of $\gamma_{\text{tot}}$ [15, 22]. In practice and especially at room temperature, the width of an emission spectrum is broader than this value. The spectrum collected from an ensemble of emitters can broaden due to the spectral broadening of each individual emitter, or because the individual emitters emit at different center frequencies as illustrated in Fig. 1.2(a) and (b). The first case is referred to as homogeneous broadening, the second case is called inhomogeneous broadening.

The light sources used in this thesis are dye molecules which form a four-level system. Figure 1.2(c) provides a schematic illustration of the decay processes in such a four-level system. An excited four-level emitter relaxes to the lowest level of the excited state by non-radiative transitions from which it decays (partly) radiatively to a vibrational sublevel of the ground state. The presence of these vibrational sublevels of the ground state are the main broadening mechanism for these type of emitters [23, 24]. Although the lifetime of the lowest excited state is relatively long (order of nanoseconds) and would give rise to a narrow spectrum ($\Delta \lambda \sim 1$ nm), the vibrational sublevels enable each molecule to undergo radiative transitions with widely varying energies leading to a broader spectrum. Typical four-level emitters therefore exhibit a long lifetime and a homogeneously broadened frequency spectrum ($\Delta \lambda \sim 50$ nm).

1.1.4 Stimulated emission, absorption, and elastic scattering

Just as vacuum fluctuations of the electromagnetic field modes are able to induce radiation from an atomic or molecular dipole, so are higher energy eigenstates of the electromagnetic modes with the same frequency. We then speak of stimulated transitions and in contrast to spontaneous emission, these transitions are not intrinsically quantum mechanical, they are as accurately described in a semiclassical framework. In the semiclassical picture, the incident field creates an oscillating dipole in the atom, which originates from the superposition of two atomic eigenstates $|\psi_1\rangle$ and $|\psi_2\rangle$ with different energies, $E_2 > E_1$. As a consequence the oscillating dipole starts radiating itself and this radiation can either be in-phase or out-phase with the incident radiation. A collection of in-phase oscillating dipoles interferes constructively with the incident field leading to an enhancement of the field, whereas a collection of out-phase dipoles interferes destructively and leads to an attenuation of the incident field [15]. These coherent processes are referred to as stimulated emission for the in-phase case and stimulated absorption for the out-phase case, yet they stem from the same dipole excitation and therefore have equivalent transition probabilities. Stimulated absorption lets the atomic system end up in the energetically excited state $|\psi_2\rangle$, while stimulated emission let the atomic system end up in the energetically lower state $|\psi_1\rangle$. When a population of atoms is considered with $N_1$ atoms in the ground state and $N_2$ atoms in the excited state stimulated emission and absorption occur simultaneously. If $N_1$ initially exceeds $N_2$ incident radiation is attenuated, if however the situation is reversed radiation is amplified. In the latter case of amplification, the population is said to be inverted.

When either of these two processes occur the original phase of the field is preserved by the oscillating dipoles of the atoms or molecules. The oscillating dipoles can easily loose their phase information however by non-radiative transitions, collisions, and spontaneous emission. This process is called dephasing and for the experiments described in this thesis, dephasing happens on order of magnitude shorter time scales than the stimulated radiative processes. Due to dephasing, the emission from a collection of in-phase excited molecules will be due to incoherent spontaneous emission.
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The change in energy level of the atom clearly distinguishes stimulated transitions from elastic scattering processes. Elastic scattering is also due to oscillating dipoles that are induced by an electromagnetic field. However, in the case of elastic scattering the energy of the particle does not change. In order to describe scattering in terms of energy states, the particle is said to be excited by the incident light to a virtual energy state from which the particle immediately falls back to its real state while reradiating. Since the atom never populates a higher energy state in reality, dephasing does not occur and the reradiated light is in phase with the incident light. The difference between scattering, stimulated emission, stimulated absorption, and spontaneous emission is summarized in Fig. 1.3. For closely isotropically packed scattering atoms in a dielectric (separation distance $\ll \lambda$) incident light is therefore hardly affected apart from obtaining a different speed, which can be derived from Maxwell’s equations by considering a homogeneous polarizability [3]. When scatterers are randomly positioned in a medium with separation distances on the order of a wavelength or longer, the propagation of waves is drastically altered. The propagation of waves through such an inhomogeneous, scattering medium is the subject of Sec. 1.2, but we will first discuss how stimulated emission is utilized in a laser, one of the most important contemporary artificial sources of radiation.

1.1.5 Lasers

The term laser is an acronym for “Light Amplification by Stimulated Emission of Radiation”. The idea of amplifying radiation by stimulated emission was originally conceived and put to practice in the microwave part of the electromagnetic spectrum [25]. Theodore Maiman built the first operating laser for visible light in 1960 [26] using a flash lamp and a ruby crystal as an amplifying medium.

Although lasers vary widely in design and operation, they all consist of three essential ingredients: a gain medium that leads to amplification of radiation, a spectrally selective feedback mechanism that enhances the interaction between light of a certain frequency and the gain medium, and a pump that provides the energy for the excitation of the gain medium.

In practice, most conventional lasers have a gain medium that consists of a collection of atoms or molecules that is population inverted [27] and a cavity consisting of high-quality
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Figure 1.4: (a) Illustration of a conventional laser. A pump mechanism (small black arrows) is used to create a population inversion in the gain medium (gray block). Two mirrors form a cavity which provides feedback for its modes. Light is amplified by stimulated emission in the gain medium. A small part of the amplified radiation is transmitted through one of the mirrors. (b) The output power of a laser for two different values of the spontaneous emission factor $\beta$. For $\beta = 1$ (gray line) there is no threshold in the output power. For $\beta < 1$ (black line) a threshold can be defined by the crossing of the extrapolated above threshold output power (dotted line) and the $x$-axis as indicated by the black dot.

The dynamics of the number of molecules in the upper laser level and the energy in the cavity mode are related. For a single-mode laser with a unit quantum efficiency four-level gain medium, the number of photons $q$ in the cavity mode and the number of molecules in the upper laser level $N$ are given by [15]

\[
\frac{dq}{dt} = -\gamma_c q + \beta N \gamma_r q + \beta N \gamma_r,
\]

\[
\frac{dN}{dt} = R - \beta N \gamma_r q - N \gamma_r,
\]

with steady-state solution

\[
q = \frac{1}{2\beta} + \frac{R}{2\gamma_c} + \frac{1}{2} \sqrt{\left( \frac{1}{\beta} - \frac{R}{\gamma_c} \right)^2 + 4 \frac{R}{\gamma_c}}.
\]

Here $\gamma_c$ is the cavity decay rate, $R$ is the pump rate, and $\beta$ is the spontaneous emission factor, which describes what the probability is that a spontaneously emitted photon ends up in the cavity mode. For a conventional laser this $\beta$-factor lies between $10^{-7}$ and $10^{-10}$ [15]. Spontaneous emission is essential in most lasers, since it provides the seed for amplification.

A plot of the steady-state solution to rate Eqs. (1.20)-(1.21) in Fig. 1.4(b) reveals that the number of photons is strongly dependent on the pump rate when $\beta < 1$. A pump threshold can be defined, $R_{\text{th}} = (1/\beta - 1)\gamma_c$, above which the number of photons starts to increase rapidly as a function of pump power. At threshold the gain in the system compensates for the losses in the cavity mode. Above threshold, stimulated emission has become the most important source of radiation and the laser mode thereby prevents that excited molecules “loose” their energy to unwanted spontaneous emission outside the cavity mode. Since radiation induced by stimulated emission is in phase with the incident radiation, the laser light can become highly coherent in both time and space. For the peculiar case of $\beta = 1$, the situation is different. Every emitted photon ends up in the cavity mode by definition and therefore the threshold in the output power disappears.
1.2 Random photonic media

The strive for perfectly ordered and clean structures has been deeply entrenched in optics. Scattering due to structural inhomogeneities or particles is indeed detrimental to many optical devices, such as glass lenses, metallic mirrors, and conventional lasers, and therefore preferably avoided. In our encounter with nature, however, scattering of electromagnetic radiation is all around us and simply cannot be neglected. Not surprisingly, the research on multiple light scattering has long been dominated by astrophysicists for whom knowledge of electromagnetic wave scattering is needed to uncover essential information about stellar systems [11, 28].

Despite the ongoing experimental efforts to fabricate nearly perfectly ordered photonic structures, it is inevitable that any man-made structure contains disorder to some extent. This unavoidable presence of disorder limits the performance of, e.g., photonic crystals whose design is based intrinsically on order [29, 30]. The exciting promises of the field of metamaterials such as cloaking and negative index materials are also based on the assumption of perfect order, and it remains to be seen whether these promises are feasible when some disorder is taken into account [31, 32].

Instead of taking order as a starting point of science and technology, one might as well accept the presence of disorder from the beginning and try to use it to one’s advantage. This idea of perceiving disorder as a strength rather than a weakness, has become more widespread in recent years. Introducing disorder by design has enabled a strong interaction between single photons and single quantum emitters via Anderson localized modes [33] and has led to the observation of transverse Anderson localization [34]. In the microwave regime, the time-reversal of scattered waves originating from a point source was shown to lead to focussing beyond the diffraction limit [35]. In optics, where time-reversal of waves is far from trivial, it was shown that manipulating the amplitude and phase of a wavefront by spatial light modulators and appropriate feedback algorithms can lead to sharp focussing outside [36, 37] and inside [38] a scattering medium. This wavefront shaping technique has been further developed and in combination with high refractive index scattering media it allows for sub-100 nm microscopy [39]. Using the technique of wavefront shaping the transmission matrix describing the transmission of light through random media was determined, allowing for the imaging of objects through opaque media [40].

Besides the development of these revolutionary disorder based technologies, the physics of wave transport through disordered systems is exciting and worth studying in itself. A better understanding of the propagation of waves in multiple scattering media explains the appearance of many objects surrounding us and the transition from metals to insulators. Rather than solving Maxwell’s equations by brute force as for example done by Finite-Difference Time-Domain calculations (FDTD) [41], which in principle would lead to precise predictions of light transport in random media, we prefer to seek accurate analytical descriptions by reducing the complexity of the system from the start. In this section we first study how single particles scatter light in Subsec. 1.2.1. The transport mean free path is introduced as the essential quantity for describing multiple scattering of light in Subsec. 1.2.2. We then discuss how both a particle and wave like description of electromagnetic radiation passing through a collection of single scatterers lead to diffusion in Subsec. 1.2.3 and Subsec. 1.2.4, and finally how the incorporation of interference leads to Anderson localization in Subsec. 1.2.5.
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Figure 1.5: The size of a scatterer determines its scattering function. (a) For particles much smaller than the wavelength the scattering is governed by Rayleigh scattering. The scattering is isotropic in the plane perpendicular to the polarization. (b) Particles with a size on the order of the wavelength are in the Mie-regime. The scattering function is very dependent on angle. (c) When the size of the particle is much larger than the wavelength ray optics can be used to describe the scattering process.

1.2.1 Single scattering

The way scattering from a particle is described depends on its size with respect to the wavelength and the refractive index contrast between the particle and the surrounding medium. We define the size parameter \( x \equiv k a \) with \( a \) the characteristic length of the particle and \( k \equiv \omega / c \) the wave number of the wave. For spherical particles \( a \) is given by the radius. The three regimes of single particle scattering are illustrated in Fig. 1.5 and explained below. When a scattering particle is much smaller than the wavelength \( (x < 1) \), the particle is said to be in the Rayleigh regime and can be treated as a dipole for which we have already calculated in Sec. 1.1.1 that the radiation pattern is isotropic in the plane perpendicular to the orientation of the dipole. The polarizability \( \alpha \) of the particle \(^2\) determines the strength of the induced dipole moment by

\[
p = \alpha \mathbf{E}.
\]

The cross section of a scatterer is a quantitative measure for the strength of a scatterer. The scattering cross section \( \sigma_s \) is defined as the ratio between the power taken out of an incident wave by scattering and the intensity \( I_0 \) of the incident wave \([28]\). By the same token we can define an absorption cross section \( \sigma_a \) by considering the power taken out of an incident wave by absorption. By using Eq. (1.23), Eq. (1.17), and \( I_0 \equiv \frac{1}{2} \epsilon_0 c E_0^2 \), we find the scattering cross section of a Rayleigh scatterer

\[
\sigma_s \equiv \frac{P}{I_0} = \frac{|\alpha|^2 k^4}{6\pi \epsilon_0^2}.
\]

This relation emphasizes again that in the Rayleigh regime the scattering of particles becomes stronger for shorter wavelengths.

When particle sizes become on the order of a wavelength, the dipole approximation used for Rayleigh scatterers breaks down. One can still view the particle as a collection of dipoles and determining its scattering properties essentially comes down to adding up the contribution from every individual dipole while taking into account interactions between the dipoles themselves \([13]\). The scattered radiation will show strong fluctuations with angle due to constructive and destructive interference, and will therefore be very anisotropic.

\(^2\) In general the polarizability of a particle is a tensor. Here we only consider particles that have isotropic polarizability, in which case the tensor can be treated as a scalar.
Figure 1.6: The scattering cross section of a sphere normalized to its geometrical cross section versus size parameter on (a) a linear plot and (b) a log-log plot. The linear plot shows how resonances appear when \( x > 1 \). The logarithmic plot illustrates the \( \lambda^{-4} \) dependence of the cross section when \( x < 1 \). When \( x \gg 1 \) the scattering cross section becomes twice the geometrical cross section. The plots are calculated using Mie-theory with \( n = 1.33 \) for the sphere and \( n = 1 \) for the environment.

The exact radiation pattern and the scattering cross section of such a scatterer can only be calculated analytically for spheres and cylinders by using Mie-theory \([13, 28, 42]\). The strong dependence on wavelength for these type of scatterers results in spectral resonances of the scattering cross section as shown in Fig. 1.6 where we have plotted the calculated scattering cross section as a function of size parameter. In these plots the Mie-regime can be clearly identified by the numerous valleys and peaks. The logarithmic plot of the cross-section highlights the \( \lambda^{-4} \) dependence of the cross-section in the Rayleigh regime.

Scatterers much larger than the wavelength fall within the geometrical optics regime and can be analyzed by ray optics. For example, the scattering from rain droplets is described by this regime and enables us to explain the intricate structure of rainbows. A main somewhat paradoxical feature of large scatterers is the fact that their scattering cross section is twice their geometrical cross section \([28]\) due to the diffraction of waves at the edges of its geometrical shadow. The calculations shown in Fig. 1.6 indeed indicate that the scattering cross section becomes twice the geometrical cross section for \( x \gg 1 \).

### 1.2.2 Multiple scattering and random walks

When a system with characteristic length scale \( L \) contains more than one scatterer, waves have the possibility of scattering more than once. This probability increases when the density \( n \) of scatterers is increased or when the scattering cross section of the individual scatterers becomes larger, because the average path length between two scattering events then decreases. This length is called the scattering mean free path \( \ell_s \) and is given by

\[
\ell_s = \frac{1}{n \sigma_s}.
\]

A sample is in the multiple scattering regime when \( \ell_s < L \). In the previous section we have seen that particles do not necessarily scatter isotropically. Mie-spheres for example scatter preferentially in the forward direction. It therefore can take more than just one scattering event to completely randomize the wave’s direction. The traversed length that is required to loose all information about the input wave’s initial direction, is called the transport mean free path \( \ell \). This important quantity can be found by normalizing the scattering mean free
1.2. Random photonic media

![Figure 1.7: Random walks in (a) 1D and (b) 2D with unit step size. The light gray curves are examples of 10 random walks in 1D and 8 in 2D with 5000 and 500 steps respectively. The black curves are the ensemble averages over 100 samples. The dark gray curves indicate the root mean square of the displacement, which has been parameterized in the 2D case. The black bar equals 10 unit steps.](image)

A medium remains (partly) transparent as long as \( \ell > L \). For opaque media on the other hand the transport of waves is dominated by multiple scattering. Describing the exact wave propagation through such a complex medium obviously becomes a hopeless endeavor, which only the naivety of a computer might resolve. We shall need to introduce approximations and secondly mainly study the properties of ensemble averages, in order to not leave the physical understanding out of sight.

A simple description of wave transport in random media can be obtained by ignoring the wave character of electromagnetic radiation altogether. In this picture, the radiation is treated as a particle and the scatterers simply change the direction of these particles from one \( k \)-vector to another as if the multiple scattering medium is playing a pinball game with the wave. The radiation “particle” undergoes a random walk [43, 44] through the medium with an average step size \( \ell \). At every step \( i \) the wavevector changes direction which we indicate by a unit vector with random orientation \( \hat{k}_i \).

Figure 1.7 shows examples of random walks in 1D and 2D. By averaging over a large number \( N \) of these stochastic processes, one is able to obtain accurate approximations for the energy density in a random medium in many situations. For example, one can deduce that for isotropic random walks the mean position \( \langle r \rangle \) after \( n \) steps is the starting point by realizing

\[
\langle r(n) \rangle = \frac{1}{N} \sum_{i=1}^{N} \left[ r_i(n-1) + \ell_i \hat{k}_i \right] = \langle r(n-1) \rangle = \langle r(0) \rangle,
\]

because \( \hat{k}_i \) averages out to zero. A similar argument for the mean-square of the position yields

\[
\langle |r(n)|^2 \rangle = \frac{1}{N} \sum_{i=1}^{N} \left[ |r_i(n-1)|^2 + 2 \ell_i \hat{k}_i + \ell^2 \right] = \langle |r(n-1)|^2 \rangle + \ell^2 = n \ell^2,
\]

which shows that the average distance to the starting point scales with the square root of steps [44]. These two properties of isotropic random walks can also clearly be seen in the ensemble averaged curves in Fig. 1.7.

1.2.3 Particle diffusion

If the concentration \( C(r, t) \) of particles that undergo a random walk is not spatially uniform, the flow per unit area \( J \) of particles is towards creating spatial homogeneity. This general
property of a collection of randomly walking particles is known as Fick’s first law and is mathematically formulated as

\[ J = -D \nabla C, \quad (1.27) \]

where \( D \) is the diffusion constant which is proportional to both the step size of the random walks and the speed of the particles. In a medium where no particles are created or annihilated any change of concentration at a certain point must be due to a net flow of particles through the infinitesimal surface surrounding the point:

\[
\frac{\partial C}{\partial t} = -\nabla \cdot J, \quad (1.28) \\
\frac{\partial C}{\partial t} = D \nabla^2 C. \quad (1.29)
\]

Here we have used Eq. (1.27) to obtain Fick’s second law in Eq. (1.29), better known as the classical diffusion equation. This versatile equation is omnipresent in all exact sciences, its applications vary from describing heat conduction [45] and molecular transport in cells [43] to price fluctuations in stock markets [46]. A source term \( S \) can be incorporated by adding it up to the right hand side of Eq. (1.29). If a time-independent unit source is placed at the origin \( \delta(\mathbf{r} - \mathbf{r}') \) of an infinite medium, Eq. (1.29) reduces to the Poisson equation whose solution is given by

\[ C(\mathbf{r}) = \frac{1}{4\pi D|r|}. \]

1.2.4 Wave diffusion

Diffusion is an unavoidable phenomenon in the case of a collection of particles performing an isotropic random walk. Any random-walk-like description of radiation will therefore inevitably end up with some form of the diffusion equation after averaging over disorder. However electromagnetic radiation is a wave in the first place and we have not yet justified that Eq. (1.29) applies to wave propagation in random media. Coherent waves propagating through random media give rise to a very irregular intensity pattern known as speckle, that is caused by constructive and destructive interference at random positions. In order to understand why waves diffuse, the wave nature needs to be taken into account from the start and speckles need to be averaged out. Rather than using the vector wave Eq. (1.6) found in Sec. 1.1, we treat the waves as a scalar\(^3\) by considering the Helmholtz equation for monochromatic wave amplitude \( \Psi(\mathbf{r}, t) = \psi(\mathbf{r})e^{i\omega t} + \text{c.c.} \)

\[
\nabla^2 \Psi - \frac{\epsilon(\mathbf{r})}{c^2} \frac{\partial^2 \Psi}{\partial t^2} = 0, \quad (1.30) \\
-\nabla^2 \psi - \frac{\omega^2}{c^2} \epsilon(\mathbf{r}) \psi = 0, \quad (1.31)
\]

with \( \epsilon(\mathbf{r}) \) the dielectric constant and \( \omega \) the frequency of the wave. The solution to Eq. (1.31) depends on the variation of dielectric constant over space and our goal is to find it in a multiple scattering medium in which the dielectric constant fluctuates strongly and where a unit source is placed at the origin. In the wave equation above, it is implicitly assumed that the scatterers and the surrounding medium have a constant, but different, dielectric constant. This assumption allows us to neglect a term containing the gradient of the dielectric constant [47]. A more extensive treatment of the theory in this section can\(^3\)

\(^3\)By writing the electromagnetic waves as a scalar, the polarization of the waves is ignored.
be found in the review by Van Rossum and Nieuwenhuizen [48] and the article by Van der Mark et al. [49]. We adopt the same sign convention as Ref. [50]. For simplicity reasons, point scatterers embedded in an environment with $\epsilon = 1$ are assumed in what follows. Thus for a scatterer at position $R_j$ we have $\epsilon(r) = 1 + \mu\delta(r - R_j)$ where $\mu$ is a constant with dimension volume.

**Amplitude propagation**

Before considering the propagation of waves from a unit source in a multiple scattering medium, let us first study the propagation from a unit source in free space. The free space propagator is also known as the bare Green function and due to its importance this function receives its own symbol $g(r)$

\[
-\nabla^2 g - \frac{\omega^2}{c^2} g = \delta(r),
\]

\[
g(r) = \frac{e^{i\frac{\omega}{c}|r|}}{4\pi|r|},
\]

where the solution has been obtained after solving the first equation in Fourier space and transforming back. The solution for the wave amplitude when a collection of sources $S(r)$ is present is then simply given by the superposition principle

\[
\psi(r) = \int g(r, r')S(r')dr'.
\]

If a single point scatterer with $V(r) \equiv V\delta(r - R_j) = \mu\frac{\omega^2}{c^2}\delta(r - R_j)$ is introduced into the system at point $R_j$, one can rewrite Eq. (1.31) as $-\nabla^2 \psi - \frac{\omega^2}{c^2} \psi = V(r)\psi$. By using $V(r)\psi$ as the source term in Eq. (1.34), we find an iterative equation for the wave amplitude at point $r$

\[
\psi(r) = \psi_0(r) + \int g(r, R_j)V\delta(r' - R_j)\psi(r')dr',
\]

\[
= \psi_0(r) + g(r, R_j)V\psi(R_j),
\]

\[
= \psi_0(r) + g(r, R_j)t\psi_0(R_j).
\]

Here $\psi_0$ is the solution of the wave equation without the scatterer present, and $t$ has been introduced as part of the single particle scattering matrix $t_j$ given by

\[
t_j(r_1, r_2) = \delta(r_1 - R_j)\delta(R_j - r_2)t,
\]

\[
t = V + Vg(R_j, R_j)V + Vg(R_j, R_j)Vg(R_j, R_j)V + \ldots
\]

The scattering matrix contains the properties of a single scatterer as described in Sec. 1.2.1. For a point scatterer this $t$ as given by Eq. (1.39) is ill-defined, because the real part of $g(R_j, R_j)$ blows up. The $t$-matrix is therefore often approximated by considering only the first two terms and by replacing the infinite real part of the second term by a finite term. In the remainder of this section, we will simply describe a single scatterer by its $t$-matrix without making its form explicit.

---

4The polarizability of a real scatterer depends strongly on frequency as discussed in Sec. 1.2.1. The $t$-matrix is therefore also frequency dependent. To keep the notation concise we do not show this frequency dependence explicitly in the text.
With the description of single scattering at our disposal, let us now seek expressions for the wave amplitude inside a medium containing $N$ scatterers with a unit source at $r'$. The amplitude full Green function $G(r,r')$ is the solution to

$$ -\nabla^2 G(r,r') - \left[ \frac{\xi^2}{c^2} + \sum_{j=1}^{N} V_j(r) \right] G(r,r') = \delta(r-r'). \quad (1.40) $$

Again, this equation can be rewritten as an iterative equation

$$ G(r,r') = g(r,r') + \int g(r,r_1) \sum_{j=1}^{N} V_j(r_1) G(r_1,r') dr_1. \quad (1.41) $$

This equation considers all possible combinations of scattering from particle to particle. Some of these combinations have already been encountered in building up the $t$-matrix of a single scatterer, namely the repetitive scattering of waves from one scatterer. To simplify matters considerably, we neglect all recurrent scattering terms in Eq. (1.41): a wave does not return to the same scatterer once it has encountered another scatterer. An insightful way of visualizing this so-called Independent Scattering Approximation (ISA) is by using Feynman diagrams

$$ G_{ISA}(r,r') = - - \times - - \times - - \times - - \times - - \times - + \ldots \quad (1.42) $$

The lines indicate the bare Green function and the crosses are the $t$-matrices of the individual scatterers. Implementing the ISA in Eq. (1.41), we find

$$ G_{ISA}(r,r') = g(r,r') + \int \int g(r,r_1) \sum_{j=1}^{N} t_j(r_1,r_2) G_j(r_2,r') dr_1 dr_2, \quad (1.43) $$

$$ = g(r,r') + \sum_{j=1}^{N} g(r,R_j) tG_j(R_j,r'), \quad (1.44) $$

where $G_j$ denotes the ISA Green function of the same system excluding scatterer $j$, the replacement of $G$ with $G_j$ is a consequence of the fact that we restrict ourselves to the ISA. Repetitive scattering from one scatterer is included in the $t$-matrix.

Since the aim of this section is to find diffusion of waves, the average over disorder realizations needs to be considered which we denote by angular brackets $\langle \cdot \rangle$. The averaged Green function has to be translationally invariant $\langle G(r,r') \rangle \equiv G(r-r')$. Moreover for a scattering medium in the thermodynamic limit $G_j(r-r') = G(r-r')$, which follows from assuming that taking away a single scatterer does not alter the averaged system’s behavior significantly. The average of any function is found by integrating the function over space and dividing by the volume $V_{\text{tot}}$, thus for the $N$ scatterer functions the averaging procedure is given by $\int \prod_{j=1}^{N} \frac{dR_j}{V_{\text{tot}}}$. If this averaging over disorder procedure is applied to element $x$ of the sum in Eq. (1.44) one finds

$$ \int g(r,R_x) tG_x(R_x,r') \prod_{j=1}^{N} \frac{dR_j}{V_{\text{tot}}} = \frac{t}{V_{\text{tot}}} \int g(r-R_x) \int G_x(R_x,r') \prod_{j=1,j\neq x}^{N} \frac{dR_j}{V_{\text{tot}}} \frac{dR_x}{V_{\text{tot}}} \quad (1.45) $$

$$ = \frac{t}{V_{\text{tot}}} \int g(r-R_x) G(R_x-r') dR_x, \quad (1.46) $$
which applied to the whole sum in Eq. (1.44) results in

$$G_{\text{ISA}}(\mathbf{r} - \mathbf{r}') = g(\mathbf{r} - \mathbf{r}') + \sum_{j=1}^{N} \frac{t}{V_{\text{tot}}} \int g(\mathbf{r} - \mathbf{R}_j)G(\mathbf{R}_j - \mathbf{r}')d\mathbf{R}_j,$$

(1.47)

$$= g(\mathbf{r} - \mathbf{r}') + nt \int g(\mathbf{r} - \mathbf{R}_j)G(\mathbf{R}_j - \mathbf{r}')d\mathbf{R}_j,$$

(1.48)

where \( n = n/V_{\text{tot}} \) is the density of scatterers and all scatterers have identical \( t \)-matrices.

The remaining integral is a convolution, which suggests this equation is again easier to solve in Fourier space. In real space the solution is approximated by

$$G(\mathbf{r}) = \frac{e^{iK|r|}}{4\pi|r|},$$

(1.49)

where the effective complex wave number is given by

$$K = \sqrt{\omega^2/c^2 + nt} \equiv k_{\text{eff}} + i/(2\ell_s)$$

with \( k_{\text{eff}} = n_{\text{eff}}\omega/c \) and \( n_{\text{eff}} \) the effective refractive index.

Equation (1.49) describes how the average wave amplitude propagates through a random medium: it decays exponentially with the scattering mean free path. Yet, the exponential decay does not resemble the result obtained for the concentration in the classical diffusion equation. If we would simply study the intensity by \( \langle G \rangle \langle G^* \rangle \) only the intensity which is left in the incident radiation is described. In fact, we have derived Lambert-Beer’s law that describes how a coherent beam decays due to scattering.

**Intensity propagation**

In order to describe diffusion of waves, the averaging needs to be done at the intensity level: after the multiplication of the amplitude with its complex conjugate. To do so, let us return to the iterative Eq. (1.44) obtained in the independent scattering approximation, but instead study \( G(\mathbf{r}_1, \mathbf{r}_2)G^*(\mathbf{r}_3, \mathbf{r}_4) \)

$$G(\mathbf{r}_1, \mathbf{r}_2)G^*(\mathbf{r}_3, \mathbf{r}_4) = \left[ g(\mathbf{r}_1, \mathbf{r}_2) + \sum_{j=1}^{N} g(\mathbf{r}_1, \mathbf{r}_j)t_jG_j(\mathbf{r}_j, \mathbf{r}_1) \right] \times$$

$$\left[ g^*(\mathbf{r}_3, \mathbf{r}_4) + \sum_{j=1}^{N} g^*(\mathbf{r}_3, \mathbf{r}_j)t_jG^*_j(\mathbf{r}_j, \mathbf{r}_4) \right].$$

(1.50)

Obviously this equation becomes very complicated due to the multiplication of two sums and the iteration terms. The Feynman diagram visualizes the above equation in a more insightful manner,

$$G(\mathbf{r}_1, \mathbf{r}_2)G^*(\mathbf{r}_3, \mathbf{r}_4) = \left[ \begin{array}{c} - + -\times - + -\times -\times -\times + \ldots \end{array} \right] \times$$

$$\left[ \begin{array}{c} - + -\times - + -\times -\times -\times + \ldots \end{array} \right]^*$$

$$= \begin{array}{c} \begin{array}{c} - + -\times - + -\times -\times -\times + \ldots \end{array} \end{array}$$

(1.51)

Here multiplication of an amplitude with a complex conjugate amplitude is symbolized by writing them underneath each other with arrows pointing in opposite directions and
scattering from the same scatterer is indicated by the dotted lines. Note that recurrent scattering has again been ignored. The trick in analyzing the ensemble averaged form of this equation, lies in knowing which diagrams to consider and which ones to neglect. For example, ensemble averaging over all diagrams in which the amplitude and complex amplitude do not share any scatterer results in the coherent propagation of waves as already found in the amplitude section. The second simplest diagrams to analyze are those that have one or more scatterers in common in a sequential manner, the so-called ladder terms. If we let a triple line denote the ensemble averaged dressed Green function we obtain\(^5\)

\[
\langle L(r_1, r_2)L^*(r_3, r_4) \rangle = \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \ldots \tag{1.52}
\]

\[
= \mathcal{L} \tag{1.53}
\]

with

\[
\mathcal{L} \equiv \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \ldots \tag{1.54}
\]

\[
= \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} + \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2} \\
\text{Diagram 3} \\
\text{...}
\end{array} \mathcal{L} \tag{1.55}
\]

In real space the Bethe-Salpeter like expression for this ladder vertex reads

\[
\mathcal{L}(r) = ntt^*\delta(r) + ntt^* \int \frac{e^{-r'/\ell_s}}{(4\pi|r'|)^2} \mathcal{L}(r-r') dr'. \tag{1.56}
\]

Here for the dressed Green function Eq. (1.49) was used. Conservation of energy allows us to find a relation between the scattering cross section and the \(t\)-matrix of an individual scatterer: \(ntt^* = 4\pi/\ell_s\). With help from this relation and by transforming to Fourier space, one finally obtains

\[
\mathcal{L}(r) = \frac{4\pi}{\ell_s}\delta(r) + \frac{3}{\ell_s^2|\mathbf{r}|} \tag{1.57}
\]

The second term in this equation describes how the intensity drops off in a multiple scattering environment. If we want to know the intensity \(\langle I \rangle\) measured at point \(r\) originating from a unit source at \(r_s\), the ladder diagram needs to get connected by dressed Green functions to the source and the observation point \([51]\)

\[
\langle I(r) \rangle = \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2}
\end{array} \mathcal{L} \begin{array}{c}
\text{Diagram 1} \\
\text{Diagram 2}
\end{array}. \tag{1.58}
\]

\(^5\)Technically the ensemble averaged Green functions given by the triple lines do not contain all scatterers, because any ensemble average would then lead to a completely connected diagram. For a large number of scatterers however we can ignore this technicality and treat all dressed Green functions equally and independently.
1.2. Random photonic media

(a) (b)

Figure 1.8: Two-dimensional representation of (a) a ladder term and (b) a most crossed term connected to both a source and an observation point. Straight arrows: dressed Green functions. Dashed arrows: complex conjugate Green functions. Ensemble averaged ladder terms give rise to diffusion. Ensemble averaged cross-terms are responsible for interference effects. Intuitively this interference can be understood by realizing that when the source and the observation point overlap the path length difference between the regular path and the complex conjugate path becomes zero. Therefore, the two paths interfere constructively at the source position.

Where we have introduced $\sigma$ to denote the observation point and $\oplus$ to denote the source. Defining $r = |r_1 - r_s|$, the two short-range vertices connecting the ladder diagram to the source and the observation point are given by

$$
\int G(r_1 - r_s)G^*(r_1 - r_s)\,dr_1 = \frac{4\pi}{16\pi^2} \int e^{-r/\ell_s} \,dr = \frac{\ell_s}{4\pi}.
$$

(1.59)

By treating these short-range vertices as points and thereby letting the ladder term run from the source to the observation point, diagram (1.58) simplifies from an integral to a straightforward multiplication

$$
\langle I(r) \rangle = \left(\frac{\ell_s}{4\pi}\right)^2 \mathcal{L}(r - r_s) = \frac{3}{16\pi^2 \ell_s |r - r_s|}.
$$

(1.60)

Thus the ensemble averaged intensity’s $r$ dependence is analogous to the stationary solution of the classical diffusion equation. We conclude that the ladder vertex is responsible for the diffusion of waves. In hindsight this should not come as too big of a surprise: ladder terms represent random walks of intensity through a random medium as illustrated in Fig. 1.8(a). By ensemble averaging these random walks one describes diffusion, just as in the particle case. Solution of the time-dependent Bethe-Salpeter like Eq. (1.56) enables us to find an expression for the diffusion constant in a random medium: $D = \frac{1}{3}v_e \ell$ with $v_e$ the energy velocity [52].

1.2.5 Anderson localization

In the previous section, we have deliberately limited our discussion to those diagrams that give rise to diffusion of light, namely the ladder terms. Other diagrams are of course also present in reality and they give rise to other phenomena. Among the most studied diagrams are the so-called “most-crossed” diagrams. In these diagrams, the shared scatterers between the amplitude and complex conjugate amplitude are encountered in a time-reversed manner as shown in Fig. 1.8(b). The intensity at the observation point now becomes strongly
dependent on its relative position to the source. If the source and the observation point overlap, the amplitude and the complex amplitude following the most crossed paths always interfere constructively and thereby lead to an enhanced intensity at this particular position. By a similar reasoning the most crossed diagrams lead to an enhanced intensity in the exact backscattering direction when illuminating a finite multiple scattering sample. This interference effect is known as enhanced backscattering (EBS) or weak localization, and has been observed for a wide variety of waves [53–56]. Over the years EBS has grown from an intriguing interference effect into a technique that enables the characterization of multiple scattering samples [57–60]. In particular because the width of the EBS cone is inversely proportional to the transport mean free path.

In lower dimensions ($d = 1, 2$), random walks have unit probability of returning to the point of origin. Therefore, interference cannot be ignored when describing the transport of waves in one-or two-dimensional media. Rather than showing a diffuse behavior, the waves are exponentially localized around the source position. The absence of diffusion due to interference is known as Anderson localization, named after the man who theoretically discovered it for electron waves in 1958 [61]. In three dimensions, the situation is more complex [62]. For weakly scattering media diffusion remains a valid approximation: the probability of a wave returning to its initial position is negligible. With an increase in scattering strength, however, interference effects become more important. When the scattering mean free path is of similar magnitude as the wavelength of radiation ($k\ell_s \sim 1$ [63]), the waves scatter so strongly near the source position that interference cannot be ignored. The diffusion of waves is brought to a complete standstill and the diffusion constant becomes zero.

Observing Anderson localization in three dimensions with classical waves is extremely challenging [64]. Partly because the potential in Helmholtz Eq. (1.31) depends quadratically on frequency, in contrast to the potential for de Broglie waves that is frequency independent [11, 65]. Increasing the wavelength in a system thus does not automatically lead to a smaller value for $k\ell_s$ and a significant effort needs to be put in designing strongly scattering samples for a particular frequency range [65]. Experiments with light have additionally been hindered by the presence of absorption, which makes it difficult to make unambiguous claims on the absence of diffusion [66–68]. Recent experiments with ultrasound and metallic beads have led to convincing results that do show signatures of Anderson localization in three dimensions [69]. Together with an experiment that visualized the localized electronic states with a scanning tunneling microscope, this experiment made it possible to study the fascinating spatial structure of localized wave functions [70, 71].

1.3 Interaction between sources and random media

Multiple scattering of radiation and the science of light generation have traditionally been studied on their own. The effect of scattering on the propagation of waves generated from a far away source, is what primarily interested the astrophysicists who initiated the field of multiple scattering of light [13, 28]. For that purpose, it suffices to assume that the incident radiation is a plane wave which also conveniently simplifies the underlying mathematics. Parallel to the efforts of understanding multiple scattering, a very successful combination of science, engineering, and business has led to an incredible development in man-made

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6Fulfilling the criterion $k\ell_s \sim 1$ is by no means a guarantee for having localization effects in a sample. It should rather be seen as a quantity that indicates when localization effects are expected to become important.
1.3. Interaction between sources and random media

Figure 1.9: Illustration of a random laser. Radiation (light gray arrows) originating from a spontaneous emission event (white disks) is amplified through stimulated emission by the excited gain medium (gray area). Scatters (dark gray disks) provide feedback to the light by lengthening the path length traveled through the gain medium. The light generated by a random laser is omnidirectional, but spectrally narrow compared to the spontaneous emission spectrum.

sources of light over the past two centuries [72, 73]. First, by the introduction of electric light sources at the end of the nineteenth century, and second by the continuous innovations in the field of laser physics starting from the second half of the twentieth century. In this section, we discuss how sources embedded in multiple scattering media can give rise to exciting new phenomena. We introduce the concept of a random laser in Subsec. 1.3.1 and show how a source can give rise to infinite range correlations in Sec. 1.3.2.

1.3.1 Random lasers

Whereas in a conventional laser mirrors are used as a feedback mechanism, a random laser uses multiple scattering of light as a feedback mechanism. A cartoon of a random laser is shown in Fig. 1.9. In a random laser, a spontaneously emitted photon gets amplified by stimulated emission while at the same time its path length in the gain medium is lengthened due to the feedback provided by multiple scattering. As a consequence the emission spectrum narrows for increasing pump powers and the output power for the peak of the spectrum shows typical threshold behavior. In contrast to a conventional laser the emitted light from a random laser is omnidirectional.

The idea of generating light inside a scattering medium by stimulated emission was already thought of in 1968 by the Soviet scientist Letokhov [74]. After some initial experiments on ground laser crystals [75, 76], the field really started in the mid 1990’s due to a dispute on a paper by Lawandy et al. describing laser action from supposedly strongly scattering samples [77, 78]. The dispute centered around the question whether the samples were truly in the diffusive multiple scattering regime or whether the samples were just redirecting some amplified light by single scattering. The Nature editor dealing with the scientific correspondence, coined the term “random laser” which has been in use ever since. A random laser can either be conceived as a multiple scattering medium with gain or as a laser system with a complex cavity configuration. In fact, both well-known laser physics effects are observed with random lasers, e.g., relaxation oscillations [79], intensity fluctuations [80], and mode coupling [81], as well as prominent multiple scattering phenomena, e.g., enhanced backscattering [82] and speckle [83].

In pioneering experimental studies by Cao et al. [84, 85], narrow features were detected
in the output spectrum of ZnO random lasers. The interpretation of these narrow spectral features has turned out to be far from straightforward and the community has not yet converged to a particular explanation. Impressive theoretical efforts [86, 87] have mainly focussed on two-dimensional random laser systems. Yet, it remains to be seen how these theoretical concepts translate to three dimensional systems and how these theories can be connected directly to experimentally relevant parameters. Anderson localization [85], Fabry-Pérot resonances [88, 89], absorption induced confinement [90], and photons traveling exceptionally long light paths through the gain medium [91], are all examples of explanations put forward that illustrate the plethora of interpretations to be found in the literature. Terminology such as “coherent random lasing” and “non-resonant feedback” has become widespread [92], but often conceals the underlying physics [93]. We therefore prefer to refrain from using this kind of terminology in this thesis.

1.3.2 $C_0$-correlation

Irregular intensity patterns that appear when coherent radiation is scrambled by a static random medium, display all kinds of intensity correlations [48]. The fact that these correlations arise indicate that the speckle patterns caused by constructive and destructive interference are not as random as we might initially think. The best-studied and easiest to observe correlation, the $C_1$ also known as the “memory-effect”, describes how speckle patterns are correlated under rotation of the sample when both illumination and detection are limited to a single $k$-vector (this configuration is typically referred to as “one channel in, one channel out”). This correlation is short-ranged, meaning that it decays exponentially with rotation angle. The two other well-known correlations, the $C_2$ and $C_3$ [94, 95], describe long-range and infinite range correlations respectively. The $C_2$ shows that the total transmitted intensity is also correlated to the $k$-vector of the incident radiation (“one channel in, all channels out”) or vice versa. The $C_3$ is the optical analog of universal conductance fluctuations and describes how much the total transmitted intensity fluctuates when radiation is incident under all possible angles (“all channels in, all channels out”).

Since the study of intensity correlations was largely initiated by scientists working in the field of condensed matter physics, it is not surprising that all these three correlation refer to samples analyzed in a transmission configuration. In contrast to electromagnetic radiation, electrons cannot be created out of nothing inside a sample. In 1999, B. Shapiro [51] calculated a new type of correlation by considering intensity generated from within random media. He considered two diffusive intensities originating from the same unit source at $\mathbf{R}_s$ propagating to $\mathbf{r}$ and $\mathbf{r} + \Delta \mathbf{r}$ that share one scatterer in close proximity to the source. Diagrammatically the correlated intensity product is given by

$$\langle I_c(\mathbf{r})I_c(\mathbf{r} + \Delta \mathbf{r}) \rangle = \mathcal{L} \mathcal{L}$$ (1.61)

This diagram is evaluated by first isolating and calculating the short-range vertex $U$, so that afterwards the diffusion ladders can be treated as if emerging from the source position.
The vertex $U$ reads

$$U = \text{diagram}$$

(1.62)

If we let $R_x$ denote the position of the shared scatterer and $r_1$ and $r_2$ denote the connection points to the ladder, the number $U$ is given by

$$U = 4 \int \int G(r_1 - R_s)G^*(r_1 - R_x)G^*(R_x - R_s) \times G(r_2 - R_x)G(R_x - R_s)G^*(r_2 - R_s) dR_x dr_1 dr_2,$$

(1.63)

where the factor $4\pi/\ell$ originates from the shared scatterer at $R_x$. The integrations over $r_1$ and $r_2$ are similar and most easily handled in Fourier-space. These two integrations return

$$U = 4 \sin k_0 r \exp \left(-\frac{r}{2\ell}\right),$$

where $r = |R_s - R_x|$. Inserting these expressions in integral Eq. (1.63) gives

$$U = \frac{4\pi}{\ell} \frac{\ell^2}{16\pi^2} \int G^*(R_x - R_s)G(R_x - R_s)e^{-r/\ell} \left(\frac{\sin k_0 r}{k_0 r}\right)^2 dR_x \quad (1.64)$$

$$= \frac{\ell}{4\pi} \frac{\ell^2}{(4\pi r)^2} e^{-r/\ell} \left(\frac{\sin k_0 r}{k_0 r}\right)^2 dR_x \quad (1.65)$$

$$= \frac{\ell}{32\pi}. \quad (1.66)$$

In the last step we have approximated $\exp(-r/\ell)$ by 1, since the integrand is dominated by the region $r \ll \ell$. Using this number for the vertex $U$, expression (1.59) for the short-range objects connecting the ladders to the observation points, and the fact that an equivalent $C_0$ diagram can also be obtained by placing the shared scatterer in the outer Green functions of the diagram, the contribution of diagram (1.61) to the intensity correlation becomes

$$C_0(\Delta r) = \frac{\langle I_c(r)I_c(r + \Delta r) \rangle}{\langle I(r) \rangle \langle I(r + \Delta r) \rangle}$$

$$= 2 \frac{\ell}{32\pi k_0} \mathcal{L}(r - R_s) \mathcal{L}(r + \Delta r - R_s) \left(\frac{\ell}{4\pi}\right)^2 \mathcal{L}(r - R_s) \mathcal{L}(r + \Delta r - R_s) \left(\frac{\ell}{4\pi}\right)^4 \quad (1.67)$$

$$= \frac{\pi}{k_0 \ell}. \quad (1.68)$$

From which we conclude that scattering close to the source induces a correlation with infinite range. Such a type of correlation means that for some realizations of disorder the speckle pattern as a whole has a higher or lower intensity than the average intensity as illustrated in Fig. 1.10. This result might come as a surprise since for all configurations of disorder the intensity originates from the same source. Apparently, random media change the output power of the source.
1.4 Outline of this thesis

In chapter 2, we argue how different types of molecular light sources can lead to surprisingly different outcomes of experiments in photonics. Random lasers are shown to be great model systems for studying the consequences of using different types of sources experimentally. A new classification scheme for sources is introduced in close analogy to the source classification in electronics. The implications of this classification for the $C_0$-correlation are discussed and illustrated with a transfer matrix calculation of a source inside a one-dimensional random stack of layers with alternating refractive indices.

By extruding such a one-dimensional system along one direction, the dynamics of Anderson localization can be studied. In chapter 3, we explain how this so-called transverse localization scheme maps a spatial coordinate onto time. Using a microwave setup, we are able to measure when the ensemble averaged wave functions localize. An eigenmode analysis of the system’s Hamiltonian is performed, in order to explain the results.

In chapter 4, diffusion inside an amplifying random medium is studied. By making use of a side-imaging technique, the random lasing threshold becomes visual in space. The system is analyzed numerically by coupling the time-dependent diffusion equation to the time-dependent equation for the population inversion. Both experimentally and numerically, we observe that gain leads to an expansion of the diffusive volume.

This expansion in volume can cause remarkable spectral effects in a random laser as illustrated by experimental studies in chapter 5. By carefully controlling the absorption of light emitted by a random laser, we are able to shift the output spectrum of a random laser by several line widths. This tunability is analyzed by describing the random laser emission spectrum with one effective cavity decay rate.

The narrow spectral features that have dominated the field of random laser are studied in chapter 6. We introduce new ways of studying these features systematically. First, the spatial structure belonging to a certain spectral feature is measured using a spectrally and spatially selective detection apparatus. Second, a wide range of random laser samples is studied to find out how the transport mean free path and the gain length influence the appearance of spikes. A conceptual model is introduced based on two laser modes that explains the results qualitatively.

Finally, in chapter 7 we conclude and put forward several subjects and applications naturally following from our research.