ESPCI PARIS | PSL

3ème année

Waves in Complex Media

Lecture notes

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Foreword

These lecture notes summarize the basic concepts and the technical developments of the courses *Waves in Complex Media* given at ESPCI Paris-PSL and at Institut d'Optique Graduate School. They do not include the many examples and applications that are discussed all along the live lecture.

The lecture assumes that the reader has basic knowledge in wave physics (optics, interferences and diffraction, electromagnetic waves) and in statistical physics (probablity distributions, first and second-order statistics).

Additional information is provided at the end of the document, in the form of complements, for readers wishing to explore certain aspects in greater depth.

The readers can also use the reference below, which naturally complements the presentation in these lecture notes:
R. Carminati and J.C. Schotland, *Principles of Scattering and Transport of Light* (Cambridge University Press, 2021).

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Part I

Scattering from particles

Chapter 1

Basic concepts in scattering theory

In this chapter we introduce the basic concepts used to describe the scattering of a monochromatic wave by a heterogeneous medium occupying a finite volume. We deal with scalar waves, that are used in most of the lecture. Scalar waves are found, for example, in acoustics or quantum mechanics. They also provide an approximate description of optical phenomena when the influence of polarization can be neglected.

1.1 Formal description of a scattering problem

We consider the scattering of a monochromatic wave with complex amplitude $E_0(\mathbf{r})$ and frequency ω, of the form $E_0(\mathbf{r},t) = \mathrm{Re}[E_0(\mathbf{r})\ \exp(-i\omega t)]$, incident on a heterogeneous medium, as sketched in Fig. 1.1.

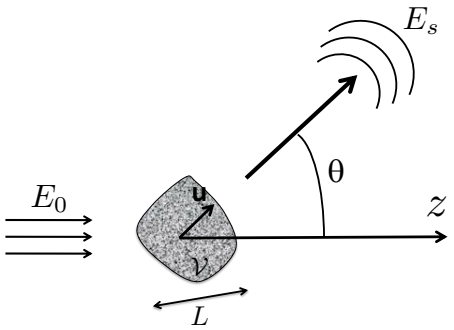

Figure 1.1: Geometry of the scattering problem. A heterogeneous material (scattering medium) is enclosed in a volume $\mathcal{V}$ with typical size L.

The scattering medium fills a finite volume $\mathcal{V}$ (with typical size L) and is described by its dielectric function $\epsilon(\mathbf{r})$ that can be complex valued, with the imaginary part describing absorption.

1.1.1 Scattered field

The complex amplitude $E(\mathbf{r})$ of the total field (the field in the presence of the scattering medium) obeys the Helmholtz equation

$$\nabla^2 E(\mathbf{r}) + \epsilon(\mathbf{r})\, k_0^2 E(\mathbf{r}) = -s(\mathbf{r})\,, \tag{1.1}$$

where $s(\mathbf{r})$ is the source of the incident field (the minus sign being chosen for later convenience) and $k_0 = \omega/c = 2\pi/\lambda$ with λ the wavelength in free space. The incident field obeys the Helmholtz equation in free space

$$\nabla^2 E_0(\mathbf{r}) + k_0^2 E_0(\mathbf{r}) = -s(\mathbf{r})\ . \tag{1.2}$$

Defining the scattered field as $E_s = E - E_0$, we immediately find that it satisfies

$$\nabla^2 E_s(\mathbf{r}) + k_0^2 E_s(\mathbf{r}) = -k_0^2[\epsilon(\mathbf{r}) - 1]\, E(\mathbf{r})\ . \tag{1.3}$$

To simplify the notations, we introduce the scattering potential $V(\mathbf{r}) = k_0^2[\epsilon(\mathbf{r}) - 1]$, and rewrite Eq. (1.3) as

$$\nabla^2 E_s(\mathbf{r}) + k_0^2 E_s(\mathbf{r}) = -V(\mathbf{r})\, E(\mathbf{r})\,. \tag{1.4}$$

Using $V(\mathbf{r})$ makes the formalism suitable for the description of different kinds of waves.

1.1.2 Integral equation

We will now show that the solution to Eq. (1.4) obeys an integral equation. To proceed, we introduce the free-space Green function G_0 that satisfies the Helmholtz equation with a delta-function source term:

$$\nabla^2 G_0(\mathbf{r}, \mathbf{r}') + k_0^2\, G_0(\mathbf{r}, \mathbf{r}') = -\delta(\mathbf{r} - \mathbf{r}')\,. \tag{1.5}$$

In three dimensions, the solution satisfying the outgoing wave condition is[1]

$$G_0(\mathbf{r}, \mathbf{r}') = \frac{\exp(ik_0 R)}{4\pi R} \tag{1.6}$$

[1] It is common to choose the outgoing Green function, which corresponds to the retarded solution in the time domain.

with $R = |\mathbf{r} - \mathbf{r}'|$, and represents a diverging spherical wave centered at $\mathbf{r}'$. Using the Green function, the incident field can be written as

$$E_0(\mathbf{r}) = \int G_0(\mathbf{r}, \mathbf{r}')\, s(\mathbf{r}')\, d^3r' \,. \tag{1.7}$$

This expression can be understood intuitively as a linear superposition of the radiation emitted by each point of the source.[2] The fact that $E_0(\mathbf{r})$ given by (1.7) is a solution to the Helmholtz equation can be checked by inserting (1.7) into (1.2) and by making use of Eq. (1.5). Likewise, the solution to Eq. (1.4) can be written

$$E_s(\mathbf{r}) = \int G_0(\mathbf{r}, \mathbf{r}')\, V(\mathbf{r}')\, E(\mathbf{r}')\, d^3r' \,. \tag{1.8}$$

The total field is obtained by superposition:

$$E(\mathbf{r}) = E_0(\mathbf{r}) + \int G_0(\mathbf{r}, \mathbf{r}')\, V(\mathbf{r}')\, E(\mathbf{r}')\, d^3r' \,. \tag{1.9}$$

This integral equation satisfied by the total field $E(\mathbf{r})$ is known as the Lippmann-Schwinger equation.

1.1.3 Far-field asymptotics

At large distance from the scattering volume, the expression of the scattered field can be simplified using the far-field approximation. For a large observation distance $r = |\mathbf{r}|$, we can use the expansion

$$|\mathbf{r} - \mathbf{r}'| \simeq r - \mathbf{u} \cdot \mathbf{r}' \,, \tag{1.10}$$

where $\mathbf{u} = \mathbf{r}/r$ is the unit vector defining the observation direction (see Fig. 1.1). This leads to the following first-order asymptotic expansion of the Green function

$$G_0(\mathbf{r}, \mathbf{r}') \simeq \frac{\exp(ik_0 r)}{4\pi r}\, \exp(-ik_0 \mathbf{u} \cdot \mathbf{r}') \,, \tag{1.11}$$

which is valid provided that the conditions $r \gg L$ and $r \gg L^2/\lambda$ hold. Using this expansion in Eq. (1.8), we find that the scattered field in the far zone takes the form

$$E_s(\mathbf{r}) = A(\mathbf{u})\, \frac{\exp(ik_0 r)}{r} \,, \tag{1.12}$$

with $A(\mathbf{u})$ the scattering amplitude given by

$$A(\mathbf{u}) = \frac{1}{4\pi} \int \exp(-ik_0 \mathbf{u} \cdot \mathbf{r}')\, V(\mathbf{r}')\, E(\mathbf{r}')\, d^3r' \,. \tag{1.13}$$

[2] Here we favor an intuitive approach, with the Green's function seen as a linear impulse response. Equation (1.7) can be established formally using the second Green identity, see for example [1] (chap. 5) or [4].

When the incident field is a plane wave with complex amplitude $E_0(\mathbf{r}) = A_0 \exp(ik_0 \mathbf{u}_{inc} \cdot \mathbf{r})$, with $\mathbf{u}_{inc}$ a unit vector defining the direction of the incident wave, it is useful to introduce the normalized scattering amplitude $\mathcal{S}(\mathbf{u}) = A(\mathbf{u})/A_0$, and write the scattered field in the far zone as

$$E_s(\mathbf{r}) = \mathcal{S}(\mathbf{u})\, A_0\, \frac{\exp(ik_0 r)}{r}\,. \tag{1.14}$$

The scattering amplitude $\mathcal{S}(\mathbf{u})$ defined this way is independent of the amplitude of the incident plane wave.

1.2 Energy conservation and optical theorem

1.2.1 Energy current and absorbed power

We assume that the scattering medium fills a volume $\mathcal{V}$ enclosed by a surface $\mathcal{S}$. Using Eq. (1.1) in volume $\mathcal{V}$ (with $s(\mathbf{r}) = 0$ since the source is outside the scattering volume), we can show that

$$E^*\, \nabla^2 E - E\, \nabla^2 E^* = -k_0^2(\epsilon - \epsilon^*)\, |E|^2\,, \tag{1.15}$$

which can also be written[3]

$$\nabla \cdot [E^*\, \nabla E - E\, \nabla E^*] + 2i\, \mathrm{Im}V\, |E|^2 = 0\,. \tag{1.16}$$

This equation takes the form of a conservation law. Let us define the energy current $\mathbf{J}$ by

$$\mathbf{J} = \frac{1}{2ik_0}\, [E^* \nabla E - E \nabla E^*]\,, \tag{1.17}$$

where the normalization is chosen in order to have $|\mathbf{J}| = |E|^2$ for a plane wave. The energy current plays the same role for scalar wave as the Poynting vector for electromagnetic waves (note the similarity with the definition of the probability current in quantum mechanics). The conservation law becomes

$$\nabla \cdot \mathbf{J} + \frac{1}{k_0} \mathrm{Im}V\, |E|^2 = 0\,. \tag{1.18}$$

Integrating over $\mathcal{V}$, and making use of the divergence theorem to transform the first integral into a surface integral, we find that

$$\int_{\mathcal{S}} \mathbf{J} \cdot \mathbf{n}\, d^2 r + \frac{1}{k_0} \int_{\mathcal{V}} \mathrm{Im}V\, |E|^2\, d^3 r = 0\,, \tag{1.19}$$

[3] We make use of the identity $\nabla \cdot [E^*\, \nabla E - E\, \nabla E^*] = E^*\, \nabla^2 E - E\, \nabla^2 E^*$.

where $\mathbf{n}$ is the outward normal on $\mathcal{S}$. The first term is the energy flux carried by the field through the surface $\mathcal{S}$. The second term correspond to the power lost by absorption within volume $\mathcal{V}$. We deduce that the absorbed power is

$$P_a = \frac{1}{k_0} \int_{\mathcal{V}} \mathrm{Im}V \, |E|^2 \, d^3r \,, \tag{1.20}$$

which, as expected, vanishes for a real potential (remember that for electromagnetic waves, a material with a real dielectric function is non absorbing).

1.2.2 Energy conservation in a scattering problem

We now derive an energy balance suitable for a scattering problem. Considering here the scattered field E_s, and using Eq. (1.4), we can show that

$$E_s^* \, \nabla^2 E_s - E_s \, \nabla^2 E_s^* = -2i \, \mathrm{Im}[V E_s^* E] \,. \tag{1.21}$$

Proceeding as above, but with the energy current of the scattered field

$$\mathbf{J}_s = \frac{1}{2ik_0} \left[E_s^* \nabla E_s - E_s \nabla E_s^* \right] \,, \tag{1.22}$$

we obtain

$$\nabla \cdot \mathbf{J}_s = -\frac{1}{k_0} \mathrm{Im}[V E_s^* E] \,. \tag{1.23}$$

Integrating over volume $\mathcal{V}$, and noting that

$$P_s = \int_{\mathcal{S}} \mathbf{J}_s \cdot \mathbf{n} \, d^2r \tag{1.24}$$

is the power carried by the scattered field (simply denoted by scattered power), we find that

$$P_s = -\frac{1}{k_0} \int_{\mathcal{V}} \mathrm{Im}[V E_s^* E] \, d^3r \,. \tag{1.25}$$

Since $E_s = E - E_0$, the integrand can be written $\mathrm{Im}[V E_s^* E] = \mathrm{Im}V|E|^2 - \mathrm{Im}[V E_0^* E]$. Inserting this splitting into Eq. (1.25), and making use of Eq. (1.20), we obtain

$$P_e = P_s + P_a \tag{1.26}$$

where

$$P_e = \frac{1}{k_0} \int_{\mathcal{V}} \mathrm{Im}[V E_0^* E] \, d^3r \tag{1.27}$$

is known as the extinguished power. Equation (1.26) is the energy balance in a scattering problem. Physically, the extinguished power corresponds to the power transferred from the incident field to the scattering medium. This power is either scattered to the far field or absorbed within the medium. Scattering and absorption both contribute to the extinction of the incident wave.

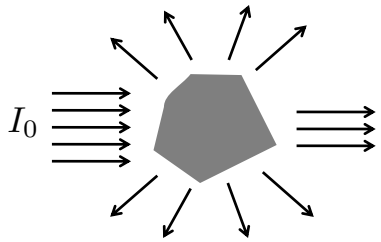

Figure 1.2: Schematic representation of extinction of an incident beam by a scattering object.

1.2.3 Optical theorem

We will now show that the extinguished power can be deduced from the scattering amplitude in the forward direction (in the direction of the incident plane wave). This result, known as the optical theorem, is a very important theorem in scattering theory.

Writing the complex amplitude of the incident plane wave as $E_0(\mathbf{r}) = A_0 \exp(ik_0 \mathbf{u}_{inc} \cdot \mathbf{r})$, Eq. (1.27) becomes

$$P_e = \frac{1}{k_0} \mathrm{Im} \left[A_0^* \int_{\mathcal{V}} \exp(-ik_0 \mathbf{u}_{inc} \cdot \mathbf{r})\, V(\mathbf{r}) E(\mathbf{r})\, d^3 r \right] . \tag{1.28}$$

The integral corresponds to the far-field scattering amplitude $A(\mathbf{u})$ defined in Eq. (1.13), calculated for the forward direction $\mathbf{u} = \mathbf{u}_{inc}$. Using the normalized scattering amplitude $\mathcal{S}(\mathbf{u})$, we can rewrite the extinguished power in the form

$$P_e = I_0 \frac{4\pi}{k_0} \mathrm{Im} \mathcal{S}(\mathbf{u}_{inc}) , \tag{1.29}$$

where $I_0 = |A_0|^2$ is the flux per unit surface carried by the incident wave.[4]

We have found that the extinguished power is determined by the scattering amplitude in the forward direction. Physically, extinction can be understood as resulting from the destructive interference between the incident field and the field scattered in the forward direction. The scattering amplitude $S(\mathbf{u}_{inc})$ encodes the relative amplitude and phase between both fields, and therefore encodes information about the extinction process.

1.3 Cross sections

Scattering and absorption cross sections are useful to characterize the fraction of the incident power that is scattered or absorbed.

[4] This can be seen by computing the energy current $\mathbf{J}_0$ of the incident field, and noting that $|\mathbf{J}_0| = I_0$.

Scattering

The power scattered in the direction $\mathbf{u}$ per unit solid angle is defined by

$$\frac{dP_s}{d\Omega} = \lim_{r\to\infty} \mathbf{J}_s \cdot \mathbf{u}\, r^2\,, \tag{1.30}$$

where $\mathbf{J}_s$ is the energy current of the scattered field.

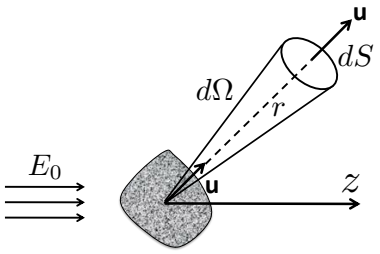

Figure 1.3: Geometry used in the defintion of the power scattered in a given direction. The surface dS supported by the solid angle $d\Omega$ at a distance r is $dS = r^2\, d\Omega$. The flux scattered in the solid angle $d\Omega$ coincides with the flux through dS.

Using the far-field expression (1.14) of E_s we find that

$$\frac{dP_s}{d\Omega} = |\mathcal{S}(\mathbf{u})|^2\, I_0\,. \tag{1.31}$$

The total scattered power is readily obtained by integrating over all directions $\mathbf{u}$, or equivalently over the solid angle Ω :

$$P_s = I_0 \int_{4\pi} |\mathcal{S}(\mathbf{u})|^2\, d\Omega\,. \tag{1.32}$$

The scattering cross section σ_s is defined by the relation $\sigma_s\, I_0 = P_s$. Using Eq. (1.32), we find that

$$\sigma_s = \int_{4\pi} |\mathcal{S}(\mathbf{u})|^2\, d\Omega\,. \tag{1.33}$$

To characterize the power scattered in a given direction, we also introduce the differential scattering cross section $d\sigma_s/d\Omega$, defined by the relation

$$\frac{d\sigma_s}{d\Omega}\, I_0 = \frac{dP_s}{d\Omega}\,. \tag{1.34}$$

We easily see that

$$\frac{d\sigma_s}{d\Omega} = |\mathcal{S}(\mathbf{u})|^2\,. \tag{1.35}$$

The differential scattering cross section is useful to describe the anisotropy of the scattering pattern (note that even for a single spherical scatterer, the scattering pattern can be strongly anisotropic, as will be seen in the next chapter).

Absorption

Similarly, we introduce the absorption cross section σ_a such that $\sigma_a\, I_0$ is the power absorbed inside the scattering medium. For a non absorbing material we evidently have $\sigma_a = 0$.

Extinction

The power transferred by the incident field to the scattering medium is either scattered or absorbed. This power is the extinguished power P_e introduced in the previous section. An extinction cross section σ_e, such that $\sigma_e\, I_0 = P_e$, can also be defined. Energy conservation [Eq. (1.26)] imposes that

$$\sigma_e = \sigma_s + \sigma_a\,. \tag{1.36}$$

Note that $\sigma_e = \sigma_s$ for a non absorbing material.

As a consequence of Eq. (1.29), we can also write

$$\sigma_e = \frac{4\pi}{k_0}\,\mathrm{Im}\mathcal{S}(\mathbf{u}_{inc}) \tag{1.37}$$

which is the optical theorem written in terms of the extinction cross section.

Finally, it is important to point out that scattering, absorption and extinction cross sections can be very different from the actual geometrical cross section σ_{geom} of the scatterer. For a particle on resonance, we can have $\sigma_e \gg \sigma_{geom}$ as seen in the example in Fig. 1.4.

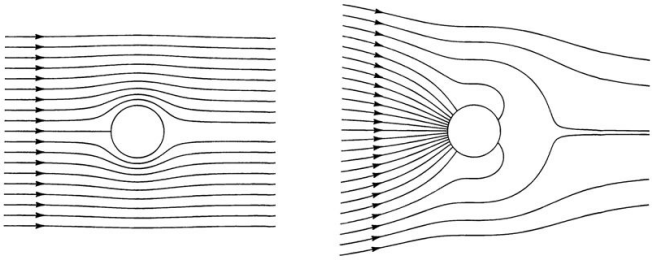

Figure 1.4: Flux lines of an optical plane wave interacting with a silver nanoparticle. The oscillation of free electrons in the metal leads to a resonance (plasmon resonance) that enhances extinction. Left: off resonance. Right: on resonance. The right figure shows a situation in which the extinction cross section is larger than the geometrical cross section. Adapted from Ref. [2].

Chapter 2

Light scattering by small particles

In this chapter we study the scattering of light by a single particle, in the framework of electromagnetic theory (this is the only chapter in which vector electromagnetic waves are considered explicitly). We discuss different interaction regimes of practical interest.

2.1 Scattering of electromagnetic waves

In this section we introduce the basic tools for the description of scattering of electromagnetic waves, along the lines developed in chapter 1.

2.1.1 Scattered field

A general scattering problem is depicted in Fig. 2.1. An external monochromatic source (frequency ω) generates the incident field, and is modelled as a localized current density $\mathbf{j}_{ext}$ located far from the scattering object (scatterer), meaning that the scatterer does not influence the source, as usually assumed in practice.

In the absence of the scatterer, the electric field produced by the external source is the incident field $\mathbf{E}_0$. In the presence of the scatterer, the total field is

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \mathbf{E}_s(\mathbf{r}) \,,$$

which defines the scattered field $\mathbf{E}_s$. The latter can be understood as the field radiated by the induced sources in the particle, the total field being the sum of the fields radiated by the external source and by the induced sources in the scatterer, which simply reflects the superposition theorem. Solving the scattering problem amounts to calculating $\mathbf{E}_s(\mathbf{r})$ in order to deduce, for example, the scattered or absorbed power.

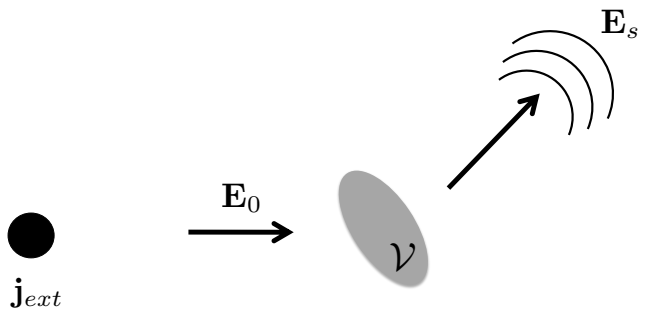

Figure 2.1: Scattering of an incident electromagnetic wave by a scattering object (denoted by particle or scatterer) with volume $\mathcal{V}$.

We describe the scatterer (assumed to be non magnetic) by its dielectric function $\epsilon(\mathbf{r})$, or refractive index $n(\mathbf{r}) = \sqrt{\epsilon(\mathbf{r})}$. The total field obeys the vector form of Helmholtz equation (easily derived from Maxwell's equation)

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \epsilon(\mathbf{r}) k_0^2 \, \mathbf{E}(\mathbf{r}) = i\mu_0 \, \omega \, \mathbf{j}_{ext}(\mathbf{r}) \tag{2.1}$$

where $k_0 = \omega/c = 2\pi/\lambda$. The incident field obeys

$$\nabla \times \nabla \times \mathbf{E}_0(\mathbf{r}) - k_0^2 \, \mathbf{E}_0(\mathbf{r}) = i\mu_0 \, \omega \, \mathbf{j}_{ext}(\mathbf{r}) \, . \tag{2.2}$$

Subtracting Eq. (2.2) to Eq. (2.1), we obtain the equation satisfied by the scattered field:

$$\nabla \times \nabla \times \mathbf{E}_s(\mathbf{r}) - k_0^2 \, \mathbf{E}_s(\mathbf{r}) = k_0^2 [\epsilon(\mathbf{r}) - 1] \mathbf{E}(\mathbf{r}) \, . \tag{2.3}$$

The term $k_0^2[\epsilon(\mathbf{r}) - 1]\mathbf{E}(\mathbf{r})$ on the right-hand side plays the role of a source term for the scattered field.

2.1.2 Green's function

As for scalar waves, the solution to Eq. (2.3) can be shown to obey an integral equation. To proceed, we need to introduce the free-space electromagnetic Green's function $\mathbf{G}_0$, which for electromagnetic waves is a second-rank tensor (the Green's function has to connect a vector source term to a vector field). This Green's function is defined as the solution to

$$\nabla \times \nabla \times \mathbf{G}_0(\mathbf{r}, \mathbf{r}') - k_0^2 \, \mathbf{G}_0(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \, \mathbf{I} \, , \tag{2.4}$$

and satisfying an outgoing wave condition when $|\mathbf{r} - \mathbf{r}'| \to \infty$ (this amounts to imposing a Green's function behaving as an outgoing spherical wave far from the source). Here $\mathbf{I}$ denotes

the unit second-rank tensor. As for scalar waves, the incident field $\mathbf{E}_0$, solution to Eq. (2.2), can be written as an integral in the form[1]

$$\mathbf{E}_0(\mathbf{r}) = i\mu_0\,\omega \int \mathbf{G}_0(\mathbf{r},\mathbf{r}')\,\mathbf{j}_{ext}(\mathbf{r}')\,d^3r'\,, \tag{2.5}$$

which describes $\mathbf{E}_0$ as the superposition of the fields radiated by each point of the source $\mathbf{j}_{ext}$.

To get some insight, let us consider the particular case of a point electric dipole source located at the point $\mathbf{r}_0$. The associated current density is $\mathbf{j}_{ext}(\mathbf{r}') = -i\omega\mathbf{p}\delta(\mathbf{r}'-\mathbf{r}_0)$, with $\mathbf{p}$ the dipole moment of the source. We find that in this case the incident field is simply

$$\mathbf{E}(\mathbf{r}) = \mu_0\,\omega^2\,\mathbf{G}_0(\mathbf{r},\mathbf{r}_0)\,\mathbf{p}\,. \tag{2.6}$$

This relation shows that the Green function $\mathbf{G}_0(\mathbf{r},\mathbf{r}_0)$ can be understood as the electric field radiated at the point $\mathbf{r}$ in free space by an elementary electric dipole located at the point $\mathbf{r}_0$.

Equation (2.6) also shows that the expression of $\mathbf{G}_0$ can be deduced from the expression of the electric field radiated by an electric dipole in free space. The electric field radiated at the point $\mathbf{r}$ by a point electric dipole $\mathbf{p}$ located at $\mathbf{r}'$ is [3]

$$\mathbf{E}(\mathbf{r}) = \frac{k_0^2}{4\pi\epsilon_0}\frac{\exp(ik_0R)}{R}\left\{\mathbf{p} - (\mathbf{p}\cdot\mathbf{u}')\mathbf{u}' - \left(\frac{1}{ik_0R} + \frac{1}{k_0^2R^2}\right)\left[\mathbf{p} - 3(\mathbf{p}\cdot\mathbf{u}')\mathbf{u}'\right]\right\} \tag{2.7}$$

with $R = |\mathbf{r}-\mathbf{r}'|$ and $\mathbf{u}' = (\mathbf{r}-\mathbf{r}')/R$. From (2.6) and (2.7) we find that

$$\mathbf{G}_0(\mathbf{r},\mathbf{r}') = \frac{\exp(ik_0R)}{4\pi R}\left[\mathbf{I} - \mathbf{u}'\otimes\mathbf{u}' - \left(\frac{1}{ik_0R} + \frac{1}{k_0^2R^2}\right)(\mathbf{I} - 3\mathbf{u}'\otimes\mathbf{u}')\right] \tag{2.8}$$

for $\mathbf{r} \neq \mathbf{r}'$. Here $\mathbf{u}'\otimes\mathbf{u}'$ is the second-rank tensor with components $[\mathbf{u}'\otimes\mathbf{u}']_{ij} = u'_iu'_j$ such that $(\mathbf{u}'\otimes\mathbf{u}')\mathbf{p} = (\mathbf{p}\cdot\mathbf{u}')\mathbf{u}'$.

In order to compute scattering amplitudes and cross sections, as for scalar waves, we will need the far-field asymptotic expression of the Green's function, that is

$$\mathbf{G}_0(\mathbf{r},\mathbf{r}') = \frac{\exp(ik_0r)}{4\pi r}\,\exp(-ik_0\mathbf{u}\cdot\mathbf{r}')\,[\mathbf{I} - \mathbf{u}\otimes\mathbf{u}] \tag{2.9}$$

with $\mathbf{u} = \mathbf{r}/r$. The validity of this approximation requires $r \gg r'$ and $r \gg r'^2/\lambda$ (far field conditions). In the far field, the Green's function is a spherical wave, corrected by a phase term that accounts for the shift in position of the source point $\mathbf{r}'$ with respect to the origin of coordinates (as for scalar waves). The main difference with the scalar Green's function in Eq. (1.11) is the presence of the tensor term $[\mathbf{I} - \mathbf{u}\otimes\mathbf{u}]$, that projects on the plane perpendicular to the obeservation direction $\mathbf{u}$ (remember that for electromagnetic waves, the electric field has to be transverse in the far field).

[1] Equation (2.5) can be established formally using the vector form of the second Green identity, see for example [1] (chap. 37) or [4].

2.1.3 Integral equation

Using the Green function $\mathbf{G}_0$, the solution to Eq. (2.3) can be written formally as

$$\mathbf{E}_s(\mathbf{r}) = k_0^2 \int \mathbf{G}_0(\mathbf{r},\mathbf{r}')\,[\epsilon(\mathbf{r}')-1]\,\mathbf{E}(\mathbf{r}')\,d^3r'\,. \tag{2.10}$$

This expression has a simple physical interpretation: denoting by $\mathbf{P}(\mathbf{r}') = \epsilon_0[\epsilon(\mathbf{r}')-1]\,\mathbf{E}(\mathbf{r}')$ the induced polarization density inside the particle, the scattered field is simply the field radiated by $\mathbf{P}$ that acts as a secondary source. Finally, the total field is obtained by adding the incident field, leading to

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + k_0^2 \int \mathbf{G}_0(\mathbf{r},\mathbf{r}')\,[\epsilon(\mathbf{r}')-1]\,\mathbf{E}(\mathbf{r}')\,d^3r'\,. \tag{2.11}$$

This integral equation is the vector form of the Lippmann-Schwinger equation (1.9) previsouly derived for scalar waves. It provides an exact description of the scattering problem. In a few particular cases (e.g. homogeneous spherical particles or particles much smaller than the wavelength) an exact analytical solution can be found. In most cases we have to rely on numerical simulations or approximate solutions.

2.1.4 Far field and scattering amplitude

The far-field expression of the scattered field is found by inserting (2.9) into (2.10). We find that $\mathbf{E}_s$ takes the form

$$\mathbf{E}_s(\mathbf{r}) = \mathbf{A}(\mathbf{u})\,\frac{\exp(ik_0 r)}{r}\,, \tag{2.12}$$

where $\mathbf{A}(\mathbf{u})$ is the vector scattering amplitude given by

$$\mathbf{A}(\mathbf{u}) = \frac{k_0^2}{4\pi}[\mathbf{I}-\mathbf{u}\otimes\mathbf{u}] \int \exp(-ik_0\mathbf{u}\cdot\mathbf{r}')\,[\epsilon(\mathbf{r}')-1]\,\mathbf{E}(\mathbf{r}')\,d^3r'\,. \tag{2.13}$$

Consider an incident plane wave propagating along direction $\mathbf{u}_{inc}$, with complex amplitude $\mathbf{E}_0(\mathbf{r}) = E_0\,\mathbf{e}_0\,\exp(ik_0\mathbf{u}_{inc}\cdot\mathbf{r})$. Here $\mathbf{e}_0$ is a unit vector defining the direction of polarization. As for scalar waves, we introduce the normalized scattering matrix $\mathbf{S}(\mathbf{u})$, a second-rank tensor such such that $\mathbf{A}(\mathbf{u}) = \mathbf{S}(\mathbf{u})E_0\,\mathbf{e}_0$. In terms of the scattering matrix the scattered field in the far zone is

$$\mathbf{E}_s(\mathbf{r}) = \mathbf{S}(\mathbf{u})E_0\,\mathbf{e}_0\,\frac{\exp(ik_0 r)}{r}\,. \tag{2.14}$$

The scattering matrix $\mathbf{S}(\mathbf{u})$ defined this way is independent of the amplitude and polarization of the incident plane wave. It is common to describe the scattering matrix for polarized light in terms of the Stokes vectors. We will not need this formalism in the lecture, but it is introduced in Complement A for the interested reader.

2.2 Optical theorem for electromagnetic waves

In this section we derive the optical theorem for electromagnetic waves. The development is similar to that used in chapter 1 for scalar waves.

2.2.1 Energy balance

With reference to the situation represented in Fig. 2.1, the incident fied satisfies the Maxwell equation

$$\nabla \times \mathbf{H}_0 = \mathbf{j}_{ext} - i\omega\epsilon_0 \mathbf{E}_0 \,. \tag{2.15}$$

In the presence of the scatterer, a polarization density $\mathbf{P} = \epsilon_0(\epsilon - 1)\,\mathbf{E}$ is created in volume V. The total field satsifies

$$\nabla \times \mathbf{H} = \mathbf{j}_{ext} - i\omega\mathbf{P} - i\omega\epsilon_0 \mathbf{E} \,. \tag{2.16}$$

By subtraction we obtain the equation satisfied by the scattered field:

$$\nabla \times \mathbf{H}_s = -i\omega\mathbf{P} - i\omega\epsilon_0 \mathbf{E}_s \,. \tag{2.17}$$

We will now write Poynting's theorem in a form involving the scattered field. Multiplying Eq. (2.17) by $\mathbf{E}_s^*$ we obtain

$$\mathbf{E}_s^* \cdot \nabla \times \mathbf{H}_s = -i\omega\mathbf{P} \cdot \mathbf{E}_s^* - i\omega\epsilon_0 |\mathbf{E}_s|^2 \,. \tag{2.18}$$

The left-hand side can be modified using the identity $\nabla \cdot (\mathbf{A} \times \mathbf{B}) = \mathbf{B} \cdot \nabla \times \mathbf{A} - \mathbf{A} \cdot \nabla \times \mathbf{B}$, leading to

$$\mathbf{H}_s \cdot \nabla \times \mathbf{E}_s^* - \nabla \cdot (\mathbf{E}_s^* \times \mathbf{H}_s) = -i\omega\mathbf{P} \cdot \mathbf{E}_s^* - i\omega\epsilon_0 |\mathbf{E}_s|^2 \,. \tag{2.19}$$

Using the Maxwell equation $\nabla \times \mathbf{E}_s = i\omega\mu_0 \mathbf{H}_s$, we get

$$-i\omega\mu_0 |\mathbf{H}_s|^2 - \nabla \cdot (\mathbf{E}_s^* \times \mathbf{H}_s) = -i\omega\mathbf{P} \cdot \mathbf{E}_s^* - i\omega\epsilon_0 |\mathbf{E}_s|^2 \,. \tag{2.20}$$

At optical frequencies, the usual observables are time-averaged powers (over a time interval much larger than $2\pi/\omega$). In complex notation, the time averaging of quadratic quantities amounts to taking $(1/2)\mathrm{Re}[...]$. Time averaging Eq. (2.20) leads to

$$\nabla \cdot \left[\frac{1}{2} \mathrm{Re}(\mathbf{E}_s^* \times \mathbf{H}_s) \right] = -\frac{\omega}{2} \,\mathrm{Im}(\mathbf{P} \cdot \mathbf{E}_s^*) \,. \tag{2.21}$$

The left-hand side is the divergence of the time-averaged Poyting vector $\mathbf{\Pi}_s$ of the scattered field. The right-hand side can be rewritten using $\mathbf{E} = \mathbf{E}_0 + \mathbf{E}_s$. We obtain the local form of the energy balance:

$$\frac{\omega}{2} \,\mathrm{Im}(\mathbf{P} \cdot \mathbf{E}_0^*) = \frac{\omega}{2} \,\mathrm{Im}(\mathbf{P} \cdot \mathbf{E}^*) + \nabla \cdot \mathbf{\Pi}_s \,. \tag{2.22}$$

In this expression, the left-hand side is the power transferred from the incident field to the scatterer per unit volume (resulting from the work done by the field on the charges inside the scatterer). The first term in the right-hand side is the power per unit volume aborbed inside the scatterer[2], and the second term is the divergence of the Poynting vector of the scattered field, that gives the power per unit surface carried by the scattered field. Integrating Eq. (2.22) over a volume enclosing the scatterer and bounded by a surface S with outward normal $\mathbf{n}$, and making use of the divergence theorem, we obtain the global energy balance:

$$P_e = P_a + P_s \tag{2.23}$$

with

$$P_e = \frac{\omega}{2}\int_V \mathrm{Im}(\mathbf{P}\cdot\mathbf{E}_0^*)\,d^3r \tag{2.24}$$

$$P_a = \frac{\omega}{2}\int_V \mathrm{Im}(\mathbf{P}\cdot\mathbf{E}^*)\,d^3r \tag{2.25}$$

$$P_s = \int_S \mathbf{\Pi}_s\cdot\mathbf{n}\,d^2r\,. \tag{2.26}$$

The extinguished power P_e is the power taken from the incident field and transferred to the scatterer. This power is either scattered (or equivalently radiated in the far field), as described by P_s, or absorbed in the scatterer, as described by P_a.

2.2.2 Extinguished power

When the incident field is a monochromatic plane wave with complex amplitude $\mathbf{E}_0(\mathbf{r}) = E_0\,\mathbf{e}_0\,\exp(i\mathbf{k}_{inc}\cdot\mathbf{r})$, the unit vector $\mathbf{e}_0$ describing the direction of polarization, we have by definition of the extinction cross section σ_e (see chapter 1):

$$P_e = \sigma_e\,\frac{\epsilon_0\,c}{2}|E_0|^2\,.$$

The factor $I_0 = (\epsilon_0\,c/2)\,|E_0|^2$ is the power per unit surface carried by the incident wave.[3] We will show that σ_e can be written in terms of the complex amplitude of the scattered field in the forward direction (i.e. in the direction of the incident plane wave). This result is known as the optical theorem.

From Eq. (2.24) and the expression of the incident plane wave, we obtain

$$P_e = \frac{\omega}{2}\mathrm{Im}\int_V E_0^*\,\mathbf{e}_0\cdot\mathbf{P}(\mathbf{r})\,\exp(-i\mathbf{k}_{inc}\cdot\mathbf{r})\,d^3r\,. \tag{2.27}$$

[2]The absorbed power per unit volume is $\mathbf{j}\cdot\mathbf{E}$ (Joule effect), which after time averaging becomes $0.5\,\mathrm{Re}(\mathbf{j}\cdot\mathbf{E}^*)$. Using $\mathbf{j} = -i\omega\mathbf{P}$, we obtain $(\omega/2)\,\mathrm{Im}(\mathbf{P}\cdot\mathbf{E}^*)$.

[3]For electromagnetic waves, we use the Poynting vector to define rigorously the flux per unit surface of a plane wave, and find $I_0 = (\epsilon_0 c/2)|E_0|^2$.

We will now show that the integral is, up to a factor, the forward scattered field. The scattered field at point $\mathbf{r}$ is

$$\mathbf{E}_s(\mathbf{r}) = \mu_0\,\omega^2 \int_V \mathbf{G}_0(\mathbf{r},\mathbf{r}')\,\mathbf{P}(\mathbf{r}')\,d^3r'\,. \tag{2.28}$$

In the far field, for an observation along direction $\mathbf{u}$, we have (see Eq. 2.9) :

$$\mathbf{E}_s(\mathbf{r}) = \mu_0\,\omega^2\,\frac{\exp(ik_0r)}{4\pi r}\,(\mathbf{I}-\mathbf{u}\otimes\mathbf{u})\int_V \mathbf{P}(\mathbf{r}')\,\exp(-ik_0\mathbf{u}\cdot\mathbf{r}')\,d^3r' \tag{2.29}$$

where the term $(\mathbf{I}-\mathbf{u}\otimes\mathbf{u})$ is simply the projection along the plane transverse to direction $\mathbf{u}$ (in the far field the electric field is transverse). Let us now assume that we measure the far field using a polarizer, that selects the component of the electric field projected along a direction $\mathbf{e}$ (note that this direction is necessarily perpendicular to $\mathbf{u}$ since the field is transverse). The measured ampitude is

$$\mathbf{e}\cdot\mathbf{E}_s(\mathbf{r}) = \mu_0\,\omega^2\,\frac{\exp(ik_0r)}{4\pi r}\int_V \mathbf{e}\cdot\mathbf{P}(\mathbf{r}')\,\exp(-ik_0\mathbf{u}\cdot\mathbf{r}')\,d^3r'\,. \tag{2.30}$$

We now make use of the scattering matrix $\mathbf{S}(\mathbf{u})$ introduced in Eq. (2.14), and rewrite the preceding equation in the form

$$\mathbf{e}\cdot\mathbf{S}(\mathbf{u})E_0\,\mathbf{e}_0 = \frac{\mu_0\,\omega^2}{4\pi}\int_V \mathbf{e}\cdot\mathbf{P}(\mathbf{r}')\,\exp(-ik_0\mathbf{u}\cdot\mathbf{r}')\,d^3r'\,. \tag{2.31}$$

From Eqs. (2.27) and (2.31), we easily see that the extinguished power can be written in terms of the scattering matrix:

$$P_e = \frac{2\pi}{\mu_0\,\omega}\mathrm{Im}[E_0^*\,\mathbf{e}_0\cdot\mathbf{S}(\mathbf{u}_{inc})E_0\,\mathbf{e}_0]\,. \tag{2.32}$$

Using the extinction cross section, this can also be written as

$$\sigma_e = \frac{4\pi}{k_0}\,\mathrm{Im}[\mathbf{e}_0\cdot\mathbf{S}(\mathbf{u}_{inc})\mathbf{e}_0]\,. \tag{2.33}$$

This result is the optical theorem, that we already discussed in chapter 1. This theorem shows that by measuring (or calculating) the scattered amplitude in the forward direction, we can deduce the extinction of the incident wave by scattering and absorption. The fact that a power is encoded in a field amplitude is not a trivial result, and reflects the interference process between the incident and scattered wave that enters the energy balance.

2.3 Particles much smaller than the wavelength

In this section we study the particular case of spherical particles with a size much smaller than the wavelength, and made of a homogeneous material with dielectric function $\epsilon(\omega)$. Such

particles can be treated in the electric dipole approximation[4]. Their scattering properties can be decribed using an electric polarizability $\alpha(\omega)$.

2.3.1 Dipole approximation and polarizability

Consider a spherical particle with radius R, located in free space with its center at position $\mathbf{r}_0$. According to Eq. (2.11), the total field at position $\mathbf{r}$ can be written

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + k_0^2 \int_{\delta V} \mathbf{G}_0(\mathbf{r}, \mathbf{r}') \, (\epsilon - 1) \, \mathbf{E}(\mathbf{r}') \, d^3 r' \tag{2.34}$$

where δV is the volume of the small particle. In this volume, assuming that $R \ll \lambda$ with λ the wavelength of the incident wave, we can assume that the field inside the particle is uniform. This field can be determined by writing Eq. (2.34) for $\mathbf{r} = \mathbf{r}_0$, in the limit $R \to 0$. We have to take care of the fact that when $\mathbf{r} \to \mathbf{r}'$ in the integral, the Green function $\mathbf{G}_0$ is singular. Indeed, the term scaling as $|\mathbf{r} - \mathbf{r}'|^{-3}$ in the expression of $\mathbf{G}_0$ (Eq. 2.8) generates a non-integrable singularity in the real part of $\mathbf{G}_0$ when $\mathbf{r} = \mathbf{r}'$. We can write

$$\begin{aligned} \mathbf{E}(\mathbf{r}_0) &= \mathbf{E}_0(\mathbf{r}_0) + k_0^2 \, (\epsilon - 1) \int_{\delta V \to 0} \mathrm{Re}[\mathbf{G}_0(\mathbf{r}_0, \mathbf{r}')] \, \mathbf{E}(\mathbf{r}') \, d^3 r' \\ &+ i k_0^2 \, (\epsilon - 1) \, \mathrm{Im}[\mathbf{G}_0(\mathbf{r}_0, \mathbf{r}_0)] \, \mathbf{E}(\mathbf{r}_0) \, \delta V \; . \end{aligned} \tag{2.35}$$

From Eq. (2.8) it can be shown that[5]

$$\mathrm{Im}[\mathbf{G}_0(\mathbf{r}_0, \mathbf{r}_0)] = \frac{k_0}{6\pi} \mathbf{I}$$

and

$$\int_{\delta V \to 0} \mathrm{Re}[\mathbf{G}_0(\mathbf{r}_0, \mathbf{r}')] \, \mathbf{E}(\mathbf{r}') \, d^3 r' = -\frac{\mathbf{E}(\mathbf{r}_0)}{3 k_0^2} + \frac{R^2}{3} \mathbf{E}(\mathbf{r}_0) \, .$$

In the last equation the first term in the right-hand side results from the singularity of the real part of $\mathbf{G}_0$ (and is independent on the volume of the particle), and the second term results form the non-singular part. The second term is negligible when R is sufficiently small, and we will neglect it (keeping this term can increase the precision in the final expression of the polarizability, but we will not discuss these subtelties in this lecture - see for example [7]). Inserting these two results into Eq. (2.35), we end up with the expression of the field inside the particle in terms of the incident field:

$$\mathbf{E}(\mathbf{r}_0) = \frac{3}{\epsilon + 2} \left[1 - i \, 3 \delta V \, \frac{k_0^3}{6\pi} \frac{(\epsilon - 1)}{\epsilon + 2} \right]^{-1} \mathbf{E}_0(\mathbf{r}_0) \; . \tag{2.36}$$

[4] When $|\epsilon| \gg 1$, which occurs for example with some metals, we may need to go beyond the electric dipole appoximation, and describe the particle using both an electric and a magnetic dipole, see for example Ref. [5].

[5] For a detailed calculation of the Green function at $\mathbf{r} = \mathbf{r}'$, including the singular real part, see for example [1] (chap. 37) or [6]. In this lecture the results will be taken for granted.

We can observe that when $\omega \to 0$ (or $k_0 \to 0$), we have $\mathbf{E}(\mathbf{r}_0) = 3\mathbf{E}_0(\mathbf{r}_0)/(\epsilon+2)$ which is a known result in electrostatics (connecting the field inside a homogeneous sphere to the external applied field). In Eq. (2.36), the additionnal term in brackets is a dynamic correction, that account for the fact that at optical frequencies we cannot *a priori* neglect radiation from the particle, that acts as an energy loss mechanism.

Once the field inside the particle is known, we can calculate the induced dipole moment:

$$\begin{aligned} \mathbf{p} &= \int_{\delta V} \mathbf{P}(\mathbf{r})\, d^3r \\ &= \int_{\delta V} \epsilon_0(\epsilon-1)\mathbf{E}(\mathbf{r})\, d^3r \\ &\simeq \epsilon_0(\epsilon-1)\,\mathbf{E}(\mathbf{r}_0)\,\delta V \\ &= \epsilon_0\,\alpha_0(\omega)\left[1 - i\frac{k_0^3}{6\pi}\alpha_0(\omega)\right]^{-1}\mathbf{E}_0(\mathbf{r}_0) \end{aligned} \tag{2.37}$$

where we have used Eq (2.36) in the last line. By definition of the polarizability $\alpha(\omega)$, we have $\mathbf{p} = \alpha(\omega)\,\epsilon_0\,\mathbf{E}_0(\mathbf{r}_0)$. From (2.37) we immediatly end up with

$$\alpha(\omega) = \frac{\alpha_0(\omega)}{1 - i\dfrac{k_0^3}{6\pi}\alpha_0(\omega)} \quad \text{with} \quad \alpha_0(\omega) = 4\pi R^3\,\frac{\epsilon(\omega)-1}{\epsilon(\omega)+2}\,. \tag{2.38}$$

We can note that for $k_0 \to 0$ (electrostatic limit), $\alpha(\omega) = \alpha_0(\omega)$. The polarizability $\alpha_0(\omega)$ is known as the quasi-static polarizability. The different between $\alpha(\omega)$ et $\alpha_0(\omega)$ results from the mechanism of radiation in the dynamic regime ($k_0 \neq 0$). The dynamic polarizability $\alpha(\omega)$ is often said to include a "radiative correction". The correction term is proportionnal to $(k_0R)^3$, and tends to zero when $k_0R \ll 1$. We can keep in mind that for the calculation of orders of magnitude, we may use the quasi-static polarizability $\alpha_0(\omega)$ instead of the full polarizability $\alpha(\omega)$. But this approximation violates energy conservation. The denominator in the expression of $\alpha(\omega)$ in Eq. (2.38) is necessary to account for energy conservation in the scattering process.

2.3.2 Cross sections

Scattering

The incident field induces an electric dipole in the particle, with dipole moment $\mathbf{p} = \alpha(\omega)\,\epsilon_0\,\mathbf{E}_0(\mathbf{r}_0)$. The power radiated by this dipole is the scattered power P_s. Recalling the expression of the power radiated by an electric dipole [3], we can write

$$P_s = \frac{\mu_0\,\omega^4}{12\pi c}\,|\mathbf{p}|^2\,.$$

By definition of the scattering cross section σ_s, the scattered power is also $P_s = \sigma_s I_0 = \sigma_s (\epsilon_0 c/2)|\mathbf{E}_0|^2$, and we can deduce

$$\sigma_s = \frac{k_0^4}{6\pi}|\alpha(\omega)|^2 . \tag{2.39}$$

It is interesting to note that:

- In a frequency range in which $\alpha(\omega)$ can be taken constant, $\sigma_s \sim \omega^4$. This frequency dependence is a feature of scattering from small particles, known as Rayleigh scattering.
- Since when $R \to 0$ we have $\alpha(\omega) \sim R^3$, the scattering cross section of a small particle scales as $\sigma_s \sim R^6$.

Extinction

The field scattered in a direction $\mathbf{u}$ is the field radiated by the induced dipole $\mathbf{p}$ in the far field. Its expression is a classical result in electrodynamics [3]. Assuming that the particle is at the origin of coordinates ($\mathbf{r}_0 = 0$), we have

$$\mathbf{E}_s(\mathbf{r}) = \frac{k_0^2}{4\pi}\frac{\exp(ik_0 r)}{r}\,\alpha(\omega)\,\mathbf{E}_{0,\perp} \tag{2.40}$$

where $\mathbf{E}_0 = E_0\mathbf{e}_0$ is the amplitude of the incident plane wave and $\perp$ denotes the projection along a plane perpendicular to $\mathbf{u}$. Since the particle is a sphere, there is no depolarization for scattering in the forward direction. The scattering matrix for $\mathbf{u} = \mathbf{u}_{inc}$ is deduced using Eq. (2.14), which gives

$$\mathbf{S}(\mathbf{u}_{inc}) = \frac{k_0^2}{4\pi}\,\alpha(\omega)\,\mathbf{I} .$$

Making use of the optical theorem Eq. (2.33) directly leads to

$$\sigma_e = k_0\,\mathrm{Im}\,\alpha(\omega) \tag{2.41}$$

showing that the exinction cross section is given by the imaginary part of the polarizability.

Here we understand the importantce of the radiative correction in Eq. (2.38). If the particle is made of a non absorbing material at the considered frequency, the dielectric function $\epsilon(\omega)$ is real, and $\alpha_0(\omega)$ (the quasi-static polarizability) is also real. But extinction does not vanish (due to scattering) and the dynamic polarizability must have an imaginary part. For a non-absorbing material, the radiative correction produces this imaginary part that ensures energy conservation. Actually, by using $\alpha_0(\omega)$ instead of $\alpha(\omega)$, we would neglect extinction by scattering.

Absorption

The absorption cross section σ_a is readily obtained by subtraction, since by energy conservation $\sigma_e = \sigma_s + \sigma_a$. Using Eqs. (2.39) and (2.41), this leads to

$$\sigma_a = k_0 \left[\mathrm{Im}\, \alpha(\omega) - \frac{k_0^3}{6\pi} |\alpha(\omega)|^2 \right] . \tag{2.42}$$

For a non-absorbing particel $\sigma_a = 0$, and the polarizability must satisfy $\mathrm{Im}\, \alpha(\omega) = [k_0^3/(6\pi)]|\alpha(\omega)|^2$.

2.4 Particles of arbitrary size

2.4.1 Particles much larger than the wavelength

For a particle of radius R very large compared to the wavelength, the laws of geometrical optics apply. For a directional beam (plane wave) encountering the particle, it seems natural to think that the scattered or absorbed light is that corresponding to the rays intercepted by the particle, and that the extinction cross section coincides with the geometrical cross section πR^2. In fact, the extintion cross-section is twice the geometrical cross section, as a consequence of diffraction. After interception by the particle, the wavefront which continues to propagate is identical to that which would be obtained by obstructing a part of the incident plane wave by an opaque disc of radius R. This wave, which is no longer a plane wave, will diffract. Diffracted energy no longer propagates in the forward direction, thus contributing to extinction. The extinction cross section is therefore larger than πR^2. How much is the increase of the extinction cross section ? The answer is obtained qualitatively by using Babinet's theorem, which states that two complementary objects (that is, whose union gives an infinite opaque plane) produce the same diffraction pattern. The opaque disk of radius R thus produces the same quantity of diffracted light as a hole of radius R in an infinite opaque plane. In this case, the fraction of incident light that is diffracted, and therefore raised to the forward direction, is the fraction which impinges on the hole of radius R. The corresponding cross section is simply the section of the hole πR^2. In total, by combining the two effects, we obtain:

$$\sigma_e = 2\,\pi R^2 \quad \text{when} \quad R \gg \lambda\,. \tag{2.43}$$

The previous result may seem surprising, even paradoxical: A large particle raises the incident beam twice the amount of energy it catches ! In fact, it must be borne in mind that this result is obtained by assuming that the observation is in the far field (at an infinitely large distance from the particle size), largely beyond the distance where a geometrical shadow is observable. Under these conditions, any light that deviates from the forward direction, even slightly, contributes to extinction. An object of a few tens of centimeters placed in front

of a window only prevents the light it actually intercepts from entering the room. On the other hand, an object with similar size in the interstellar medium, placed between a star and a telescope on Earth, will double the amount of light removed before hiting the image plane.

2.4.2 Spherical particles of arbitrary size (Mie scattering)

A rigorous theory of scattering from homogeneous and spherical particles, known as Mie theory, is available. Given the dielectric function (or refractive index) of the material, and the radius of the particle, this theory provides analytical expressions of the scattered field in the form of infinite series that can be calculated numerically. Analytical expressions of the different cross sections and of the scattering pattern (differential scattering cross section) are also available. We can find details on the theory in textbooks (for example Ref. [2]), and user-friendly solvers are easily found online. Mie theory is an extremly convenient tool in practice, to compute the scattering properties of spherical particles.

An example of numerical calculations is shown in Fig. 2.2, for a particle with radius R et refractive index m at a given wavelength λ. The figure shows the extinction efficiency versus the dimensionless parameter $2x(m-1)$, where $x = 2\pi R/\lambda$, is the so-called size parameter. We observe a large number or resonances, whose number increases with the refractive index. These resonances are a feature of the regime of Mie scattering (one often speaks of Mie resonances). On the figure also note that the vertical axis correponds to the lower curve, the other ones being shifted for the sake of visibility. When R becomes large compared to λ, the extinction efficiency tends to 2 (and not 1). We recover the fact that in the regime $R \gg \lambda$ the extinction cross section becomes $\sigma_e = 2\pi R^2$.

Another feature of Mie scattering is that when $R \gtrsim \lambda$ the scattering pattern becomes strongly peaked in the forward direction, as shown in Fig. 2.3.

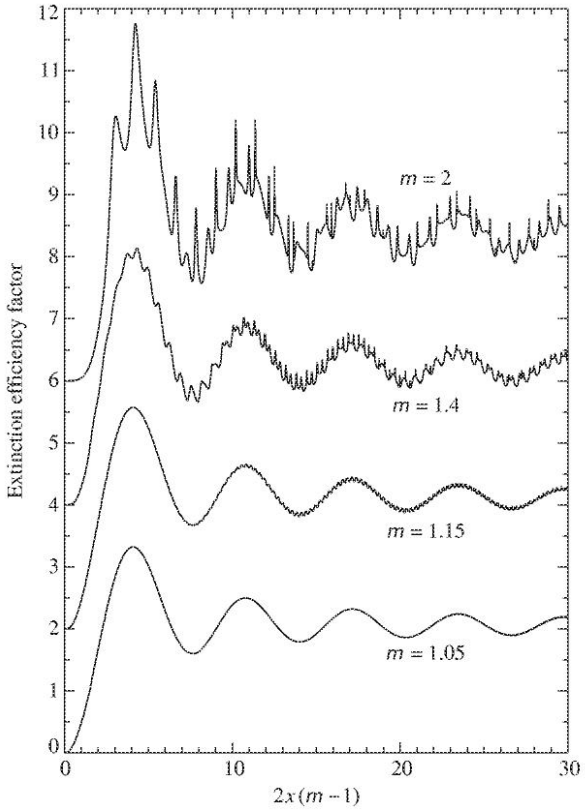

Figure 2.2: Extinction efficiency $Q_e = \sigma_e/(\pi R^2)$ of a spherical particle with radius R and refractive index m. The parameter $x = 2\pi R/\lambda$, where λ is the incident wavelength, is the size parameter. Adapted from [8].

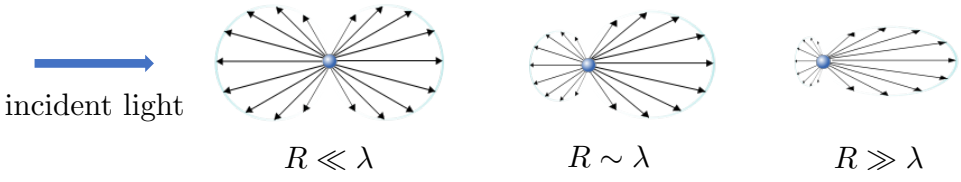

Figure 2.3: Scattering diagrams for spherical particles with different sizes. Large particles produce a strong forward scattering. Adpated from Wikipedia.

Part II

Transport in scattering media

Chapter 3

Introduction to multiple scattering

In this chapter, we introduce the framework used to describe the propagation of scalar waves in a disordered medium made of discrete scatterers. We introduce the statistical approach used all along the lecture, define the different scattering regimes, and discuss the main properties of the average field and average intensity

3.1 Scattering by an ensemble of particles

3.1.1 Born series and T matrix

We describe a scalar monochromatic wave by its complex amplitude $E(\mathbf{r})$. In chapter 1 we have shown that it obeys the Lippmann-Schwinger equation

$$E(\mathbf{r}) = E_0(\mathbf{r}) + \int G_0(\mathbf{r},\mathbf{r}')\, V(\mathbf{r}')\, E(\mathbf{r}')\, d^3r' \,, \tag{3.1}$$

with E_0 the complex amplitude of the incident wave and $V(\mathbf{r})$ the scattering potential.

In the following it will be convenient to use an operator notation, as in quantum mechanics, allowing us to rewrite Eq. (3.1) in the compact form

$$E = E_0 + G_0\, V\, E \,. \tag{3.2}$$

In this notation, G_0 and V are operators acting on the "state vector" E, such that in real space $G_0 : f \to \int G_0(\mathbf{r},\mathbf{r}')f(\mathbf{r}')d^3r'$ and $V : f \to V(\mathbf{r})f(\mathbf{r})$. Upon iterating Eq. (3.2) we find that

$$E = E_0 + G_0\, V\, E_0 + G_0\, V\, G_0\, V\, E_0 + G_0\, V\, G_0\, V\, G_0\, V\, E_0 + ... \tag{3.3}$$

which is known as the Born series. Each term in the expansion corresponds to orders of multiple scattering by the potential V.

The Born series can be factorized in the form

$$E = E_0 + G_0 \left(V + V\,G_0\,V + V\,G_0\,V\,G_0\,V + ...\right) E_0\,. \tag{3.4}$$

The series in the parenthesis defines the transition operator T (also known as T matrix)

$$T = V + VG_0V + VG_0VG_0V + ... \tag{3.5}$$

and the Lippmann-Schwinger equation can be rewritten in the form

$$E = E_0 + G_0\,T\,E_0\,. \tag{3.6}$$

We note that the field on the right is now the incident field E_0. In real space, the equation above reads

$$E(\mathbf{r}) = E_0(\mathbf{r}) + \int G_0(\mathbf{r},\mathbf{r}_1)\,T(\mathbf{r}_1,\mathbf{r}_2)\,E_0(\mathbf{r}_2)\,d^3r_1 d^3r_2\,, \tag{3.7}$$

with

$$\begin{aligned} T(\mathbf{r}_1,\mathbf{r}_2) &= V(\mathbf{r}_1)\,\delta(\mathbf{r}_1-\mathbf{r}_2) + V(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}_2)\,V(\mathbf{r}_2) \\ &+ \int V(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}')\,V(\mathbf{r}')\,G_0(\mathbf{r}',\mathbf{r}_2)\,V(\mathbf{r}_2)\,d^3r' + ... \end{aligned} \tag{3.8}$$

Handling infinite series of operators to introduce the T matrix may not seem very rigorous. We note that from Eqs. (3.2) and (3.6), one finds that the exact relation between the T matrix and the potential V is

$$T = V(I - VG_0)^{-1}\,, \tag{3.9}$$

where I is the identity operator. This is also the result obtained by summing the geometric series (3.5), showing the consistency of the approach in which infinite series are used as intermediate steps in the calculation.

With Eq. (3.6), the scattering problem has not been solved, since determining the T matrix remains as complicated as solving the integral equation (3.2). Nevertheless this formalism is well adapted to a treatment of scattering by an ensemble of particles, as we shall see.

3.1.2 Set of discrete scatterers

The potential V and the T matrix describe the scattering medium as a whole. We now assume that the medium is an ensemble of discrete scatterers in a homogeneous background. The potential can be written

$$V = \sum_j V_j \tag{3.10}$$

with V_j the potential due to scatterer number j. For example, for light scattering by a set of identical scatterers with dielectric function ϵ, we would have

$$V_j(\mathbf{r}) = k_0^2(\epsilon - 1)\,\Theta_j(\mathbf{r}) \tag{3.11}$$

where $\Theta_j(\mathbf{r}) = 1$ if $\mathbf{r}$ is inside particle number j and $\Theta_j(\mathbf{r}) = 0$ otherwise.

Using Eq. (3.10) and the defintion of the T matrix (3.5), we can write

$$T = \sum_j V_j + \sum_{j,k} V_k G_0 V_j + \sum_{j,k,l} V_l G_0 V_k G_0 V_j + ... \tag{3.12}$$

We now introduce the T matrix of a single scatterer, defined as

$$t_j = V_j + V_j G_0 V_j + V_j G_0 V_j G_0 V_j + ... \tag{3.13}$$

This operator, built in the same way as the global T matrix, fully describes scattering by a single scatterer. Using Eq. (3.13), we can rewrite the global T matrix in terms of T matrices of individual scatterers. We find that

$$T = \sum_j t_j + \sum_{j\neq k} t_j\, G_0\, t_k + \sum_{j\neq k, k\neq l} t_j\, G_0\, t_k\, G_0\, t_l + ... \tag{3.14}$$

One can check that, by inserting (3.13) into (3.14), all terms in Eq. (3.12) are recovered (and counted only once).

We have obtained an exact expression of the global T matrix in terms of T matrices of individual scatterers. The advantage of this expression is that it separates scattering by each individual particle (accounted for up to infinite order by the t_j's) from scattering between different scatterers (to increasing order in the summations).

Finally, we obtain the expression of the field by inserting Eq. (3.14) into Eq. (3.6):

$$E = E_0 + \sum_j G_0\, t_j\, E_0 + \sum_{j\neq k} G_0\, t_j\, G_0\, t_k\, E_0 + \sum_{j\neq k, k\neq l} G_0\, t_j\, G_0\, t_k\, G_0\, t_l\, E_0 + ... \tag{3.15}$$

This expression makes it possible to visualize the multiple scattering process as a set of scattering sequences between particles, involving an increasing number of scattering events. The first sum corresponds to all single scattering events. The second sum corresponds to all double scattering sequences, involving two different scatterers. The third sum corresponds to all triple scattering events (note that the first and third scatterers may be identical), etc.

3.1.3 The T matrix as a generalized polarizability

We have seen in section 2.3 that for electromagnetic waves, and for a scatterer much smaller than the wavelength, the scattering properties are determined by the polarizability $\alpha(\omega)$. This

is also true for scalar waves. In this case, the field scattered by a single scatterer located at the point $\mathbf{r}_j$ is

$$E_s(\mathbf{r}) = k_0^2 G_0(\mathbf{r}, \mathbf{r}_j)\alpha(\omega)E_0(\mathbf{r}_j)\,, \tag{3.16}$$

which is the analog of Eq. (2.40) for scalar waves. In terms of the T matrix we can also write

$$E_s(\mathbf{r}) = \int G_0(\mathbf{r}, \mathbf{r}_1)t_j(\mathbf{r}_1, \mathbf{r}_2)E_0(\mathbf{r}_2)d^3r_1 d^3r_2\,. \tag{3.17}$$

Since the above expressions must coincide, we find that the T matrix of a single subwavelength scatterer is $t_j(\mathbf{r}_1, \mathbf{r}_2) = k_0^2\,\alpha(\omega)\,\delta(\mathbf{r}_1 - \mathbf{r}_j)\delta(\mathbf{r}_2 - \mathbf{r}_j)$. For such a scatterer, the T matrix is essentially the polarizability. For an extended scatterer, the T matrix becomes a non-local operator (not reduced to a single point) and generalizes the concept of polarizability.

3.2 Statistical approach

In a disordered medium, a precise description of the detailed microstructure is out of reach. Moreover, an exact calculation of the scattered field based on a microscopic description of the medium would be useless in practice, since the observables (for example the reflectivity of a sheet of paper, or the transmissivity of a glass of milk) are averaged quantities (over space or time). These average observables usually depend on a few statistical parameters characterizing the disordered medium (for example the average number of scatterers per unit volume), and not on the precise microscopic structure. As in statistical physics, it is relevant to use a statistical approach in the first place, and deduce the statistical properties of the observables directly, without solving the full microscopic scattering problem. To proceed, we consider conceptually an ensemble of realizations of the disordered medium, and perform an ensemble averaging denoted by $\langle ... \rangle$ in this lecture. Once this statistical point of view has been adopted, with the disordered medium described as an ensemble of configurations resulting from a random process, it is common to speak of wave scattering in random media.

3.2.1 Field

The total field in one realization of the disordered medium can be written as the sum of an average value and a fluctuation:

$$E = \langle E \rangle + \delta E \quad \text{with} \quad \langle \delta E \rangle = 0\,. \tag{3.18}$$

The first term in the right-hand side is the average field (sometimes denoted by coherent field). The second term is the fluctuating field that averages to zero by definition. Since the total field is the sum of the incident and scattered fields, we can also write

$$E = E_0 + \langle E_s \rangle + \delta E_s \tag{3.19}$$

with the correspondence $\langle E \rangle = E_0 + \langle E_s \rangle$ and $\delta E = \delta E_s$.

3.2.2 Intensity

Likewise, the intensity in a specific realization of disorder can be written $I = \langle I \rangle + \delta I$. We will see that in the multiple scattering regime the average intensity $\langle I \rangle$ satisfies transport equations (such as the radiative transfer equation or the diffusion equation). Intensity fluctuations δI are central in the study of speckle which is also part of this lecture.

The average intensity is $\langle I \rangle = \langle |E|^2 \rangle$. Using Eq. (3.18), we immediatly see that

$$\langle I \rangle = |\langle E \rangle|^2 + \langle |\delta E|^2 \rangle \,. \tag{3.20}$$

The first term in the right-hand side is the power carried by the average field. It represents the so-called ballistic component I_b of the average intensity, that will be studied in the next section. The second term is the power carried by field fluctuations. Although the fluctuating field averages to zero, its average power does not vanish, and represents the diffuse component I_d of the average intensity that will be studied in the next part dedicated to transport theory. In summary, we can write

$$\langle I \rangle = I_\mathrm{b} + I_\mathrm{d}$$

with the correspondence $I_\mathrm{b} = |\langle E \rangle|^2$ and $I_\mathrm{d} = \langle |\delta E|^2 \rangle$.

3.3 Average field and ballistic intensity

In this section, we will study the behavior of the average field and of the ballistic intensity based on an intuitive phenomenological approach. A more formal approach is available, based on a closed-form equation for the average field. For the interested reader, the formal treatment is presented in Complement B. A deeper presentation can be found in textbooks and review articles [1, 9, 10, 11, 12, 13].

We consider the canonical slab geometry represented in Fig. 3.1, where a scattering medium is confined between the planes $z = 0$ and $z = L$, and illuminated from the left by a plane wave with complex amplitude E_0 and intensity $I_0 = |E_0|^2$. From Eq. (3.6), we immediately find that the average field satisfies

$$\langle E \rangle = E_0 + G_0 \langle T \rangle E_0 \,, \tag{3.21}$$

since T is the only random object in the right-hand side (the incident field E_0 is deterministic). If the medium is statistically homogeneous and isotropic, meaning that all statistical properties such as the average number of particles per unit volume are independent of position and

direction, then the average matrix $\langle T \rangle$ is homogeneous and isotropic. We can deduce that if the incident field is a plane wave, then the average field is also a plane wave propagating through an "average slab" described by effective homogeneous and isotropic properties (an effective dielectric function or effective refractive index in the case of light).

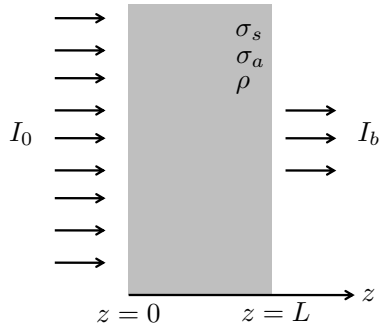

Figure 3.1: Attenuation of a collimated beam (plane wave) by a slab with thickness L filled with a statistically uniform disordered material, made of identical particles with number density ρ, and scattering and absorption cross sections σ_s and σ_a.

In order to find the behavior of the ballistic intensity $I_{\rm b} = |\langle E \rangle|^2$ transmitted through the slab, we can use a simple approach. Choosing the Oz direction to be normal to the slab interfaces, we can write an energy balance over a cylindrical volume with cross section S normal to Oz, and located between the planes z and $z + dz$:

$$I_{\rm b}(z + dz)\, S - I_{\rm b}(z)\, S = -(\rho\, S\, dz)\, (\sigma_a + \sigma_s)\, I_{\rm b}(z) = -(\rho\, S\, dz)\, \sigma_e\, I_{\rm b}(z)\ . \tag{3.22}$$

This expression describes the extinction of the ballistic intensity between z and $z + dz$, due to the particules located in the volume (their number is $\rho\, S\, dz$). We note that in this calculation it is assumed that each particle scatters as if it were alone in the medium, which is known as the *independent scattering approximation*. We can expect this approximation to be valid in diluted media. From the above we deduce the equation satisfied by $I_{\rm b}(z)$, that is

$$\frac{dI_{\rm b}(z)}{dz} + \rho\, \sigma_e\, I_{\rm b}(z) = 0$$

which after integration from $z = 0$ to $z = L$ leads to

$$I_{\rm b}(z) = I_{\rm b}(z = 0)\ \exp(-\rho\, \sigma_e\, L)\ . \tag{3.23}$$

We see that the ballistic intensity decays exponentially with the thickness L. The intensity that is lost is either redistributed in other directions by scattering (and transferred to the diffuse intensity) or absorbed. Equation (3.23) is actually the general form of the Beer-Lambert law.

For purely absorbing media ($\sigma_e = \sigma_a$), we recover the connection between the decay of the intensity and the absorbance $\rho\sigma_a L$ that is used in chemistry.

The exponential law calls for the introduction of length scales. The length $\ell_e = (\rho\,\sigma_e)^{-1}$ is the extinction mean free path (or extinction length), and the Beer-Lambert law becomes

$$I_{\rm b}(z) = I_{\rm b}(z=0)\,\exp(-L/\ell_e)\,. \tag{3.24}$$

We also define the scattering mean free path $\ell_s = (\rho\,\sigma_s)^{-1}$ and the absorption mean free path $\ell_a = (\rho\,\sigma_a)^{-1}$. Note that since $\sigma_e = \sigma_s + \sigma_a$ we have

$$\frac{1}{\ell_e} = \frac{1}{\ell_s} + \frac{1}{\ell_a}\,. \tag{3.25}$$

In this lecture we are interested in scattering materials for which the condition $\ell_a \gg \ell_s$ is satisfied.

Let us note that choosing to work with length scales is a matter of taste. One may prefer to work with attenuation coefficients (with unit m^{-1}), and define the extinction coefficient $\mu_e = \rho\sigma_e = 1/\ell_e$, the scattering coefficient $\mu_s = \rho\sigma_s = 1/\ell_s$, and the absorption coefficient $\mu_a = \rho\sigma_a = 1/\ell_a$ (these coefficients are widely used in biomedical optics).

Finally, coming back to the statistical approach, it is instructive to rewrite the ballistic intensity in the form

$$I_{\rm b} = |E_0 + \langle E_s\rangle|^2 = I_0 + |\langle E_s\rangle|^2 + 2\mathrm{Re}(E_0^*\,\langle E_s\rangle)$$

in which the last term describes the interference between the average scattered field and the incident field. Extinction by scattering and absorption imposes $I_{\rm b} < I_0$, which is made possible by this interference phenomenon (the last term in the above equation has to be negative). We can conclude that extinction is driven by the interference between the average scattered field and the incident field. We recover the physical picture of the optical theorem that was discussed in chapter 1.

3.4 Scattering regimes

We have seen that the behavior of the ballistic intensity is easy to predict (at least in a statistically homogeneous and isotropic medium, and in the independent scattering approximation). We show in this section that the Beer-Lambert law is consistent with a point of view borrowed from classical transport of particles.

3.4.1 Scattering mean free path

The scattering mean free path $\ell_s = (\rho\,\sigma_s)^{-1}$ can be understood as the average distance between successive scattering events. To see this, let us consider a non absorbing medium

($\ell_e = \ell_s$) and rewrite Eq. (3.22) in the form

$$I_{\rm b}(z+dz) \, = I_{\rm b}(z) - \frac{dz}{\ell_s} \, I_{\rm b}(z) \, . \tag{3.26}$$

Considering the intensity as a flux of classical particles (we will use the term "photons" for convenience), we can understand $I_{\rm b}(z)$ as the number of ballistic photons propagating along direction Oz, or in other words as the number of photons that have not been scattered before reaching the depth z within the medium. The above equation shows that $(dz/\ell_s) \, I_{\rm b}(z)$ is the number of photons that are scattered between z and $z+dz$. Normalizing by the number of incident photons at depth z, we can say that the probability for a photon to be scattered between z and $z+dz$ is dz/ℓ_s.

Let us now take an arbitrary photon that propagates along a direction that we choose as Oz, right after a scattering event. The probability for this photon to be scattered again after a distance z is

$$P(z) \, dz = \exp(-z/\ell_s) \, \frac{dz}{\ell_s}$$

where $P(z)$ in the left-hand side is the probability density. The average distance before the next scattering event is

$$\langle z \rangle = \int_0^{+\infty} z \, P(z) \, dz = \ell_s \, .$$

We have shown, using a point of view borrowed to the kinetic theory of classical particles, that ℓ_s is the average distance between two successive scattering events. The name "mean free path" given to ℓ_s is now clear.

3.4.2 Single and multiple scattering

Using the scattering mean free path ℓ_s, we can define three regimes for the scattering of waves in a disordered medium with characteristic size L:

- $L \ll \ell_s$: Ballistic regime (no scattering)
- $L \simeq \ell_s$: Single scattering regime
- $L \gg \ell_s$: Multiple scattering regime.

In the multiple scattering regime, the ballistic intensity is completely extinguished and energy transport occurs through the diffuse intensity.

3.4.3 Homogenization (a quick look)

The regime of homogenization is expected when all structural scales in the medium are much smaller than the wavelength. Indeed, in this case the wave does not "resolves" the microstructure of the medium and there is no scattering. The wave sees an effective homogeneous medium, and field fluctuations $\delta E \to 0$. Window glass is an exemple of a material that is disordered, but homogeneous for visible light. Homogenization is a difficult subject that we do not pretend to cover here. Instead, we only discuss an example based on scaling laws to get a first insight.

Let us assume that starting from a cloud of randomly distributed particles in a given volume, we cut the particles in smaller and smaller pieces, while keeping the volume fraction f constant (meaning that we do not remove or add material in the volume). When the size R of the particles becomes much smaller than λ, they behave as electric dipoles and we have seen that their scattering cross section scales as (see Eq. 2.39):

$$\sigma_s = \frac{\omega^4}{6\pi c^4}|\alpha(\omega)|^2 \sim R^6 \,.$$

Since the volume fraction $f = (4\,\pi R^3/3)\,\rho$ is constant, the number density ρ scales as R^{-3}. As a result, the scattering mean free path scales as

$$\ell_s = \frac{1}{\rho\,\sigma_s} \sim R^{-3} \,.$$

We see that when the size of the particles $R \to 0$, the scattering mean free path $\ell_s \to \infty$. The medium becomes less and less scattering, although the amount of material does not change. When $\ell_s \gg L$ with L the size of the medium, there is no more scattering and only a ballistic intensity is observed. This very simple example illustrates the idea of the homogenization limit.

3.5 Diffuse intensity: Towards a transport equation

To compute the full average intensity $\langle I \rangle = \langle |E|^2 \rangle$, we could in principle start from the expression (3.15) of the field E in terms of scattering sequences, write a similar expansion for E^*, and compute the product that includes all cross terms. Two scattering sequences for E and E^* are schematically represented in the left panel in Fig. 3.2. In practice, there is no hope of being able to calculate the average value of the products of pairs of different sequences. Multiple scattering theory is a framework developed to handle such an averaging process for the intensity [1, 9, 11, 13]. Reviewing multiple scattering theory is beyond the scope of this lecture. Instead, we will briefly outline the main idea that leads to a transport theory for the diffuse intensity. For the interested reader, a summary of the formal theory for the average intensity is given in Complement C.

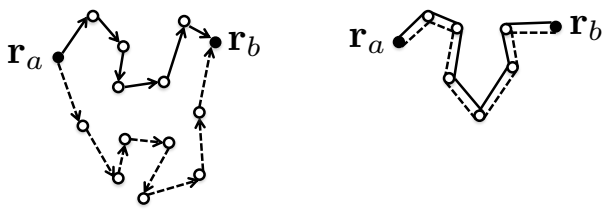

Figure 3.2: Left: Schematic representation of scattering sequences for E (solid line) and E^* (dashed line) connecting a point $\mathbf{r}_a$ to a point $\mathbf{r}_b$. White circles stand for scattering events (scatterers). Right: Contributions to the average intensity in the ladder approximation. E and E^* follow the same scattering sequences.

When the scattering mean free path ℓ_s is large compared to the wavelength λ, we can expect the product of two different scattering sequences to vanish on average.[1] Indeed, even two scattering sequences differing by only one scattering event induce a phase difference on the order of $k_0\ell_s \gg 1$. By crossing all the possibilities, we can expect to have large and small phase shifts, uniformly distributed over $[0, 2\pi]$. As a result, only the contributions resulting from two fields E and E^* following the same scattering sequences are significant in the calculation of $\langle I \rangle$, as represented in the right panel in Fig. 3.2 (the contribution of cross terms between different scattering sequences vanishes on average). Keeping only these contributions is known as the ladder approximation. A more formal presentation of the ladder approximation is given in Complement C.

We end up with the following picture: in the regime $k_0\ell_s \gg 1$, the diffuse intensity can be understood as the sum of intensities following different scattering paths. We are left with a picture in which the wave aspect can be forgotten (interferences can be neglected). The problem becomes similar to a problem of transport of classical particles. In the next two chapters, we address the transport of intensity in this setting, based on the radiative transfer equation, and on the diffusion approximation.

[1] To get an order of magnitude, consider biomedical optics. The wavelength is $\lambda \simeq 1$ μm, and the scattering mean free path in biological tissues is $\ell_s \simeq 100$ μm.

Chapter 4

Radiative Transfer Equation

In this chapter we derive a transport equation for the averaged intensity in a scattering medium, known as the radiative transfer equation (RTE). We use a phenomenological approach, based on an energy balance, that reproduces the historical derivation presented in the context of astrophysics [14]. A similar transport equation has been introduced for the transport of neutrons in nuclear reactors [15]. A derivation of the RTE starting from the wave equation is available (see for example [1], [10] or [11]), and relies on the ladder approximation briefly discussed at the end of chapter 3 .

4.1 Specific intensity

Consider an elementary surface dS with normal $\mathbf{n}$ and located at point $\mathbf{r}$, as represented in Fig. 4.1. The power carried by the wave at a frequency ω and flowing through the surface can be written

$$P(\mathbf{r},t) = dS \int_{4\pi} I(\mathbf{r},\mathbf{u},t)\,\mathbf{u}\cdot\mathbf{n}\,d\Omega \tag{4.1}$$

where $d\Omega$ means an integration over the solid angle, or equivalently over the direction defined by the unit vector $\mathbf{u}$. [1] In this expression, $I(\mathbf{r},\mathbf{u},t)$ is the specific intensity that represents the average power per unit surface flowing in direction $\mathbf{u}$ at point $\mathbf{r}$ and time t (unit W.m^{-2}.sr^{-1}).[2] Here it is assumed that the wave is quasi-monochromatic at a frequency ω (the dependence on ω is omitted to keep simple notations). The time dependence represents slow variations of the power (compared to $2\pi/\omega$), due for example to a pulse excitation.

[1] In spherical coordinates $\int_{4\pi} d\Omega = \int_0^{\pi}\int_0^{2\pi} \sin\theta\, d\theta\, d\phi$.

[2] The specific intensity plays for the radiation the same role as the Boltzmann distribution function for the transport of particles.

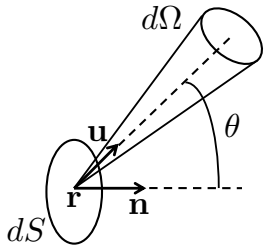

Figure 4.1: Geometry used for the definition of the specific intensity.

From the specific intensity we can define the energy current

$$\mathbf{q}(\mathbf{r},t) = \int_{4\pi} I(\mathbf{r},\mathbf{u},t)\,\mathbf{u}\,d\Omega\,, \tag{4.2}$$

that coincides (up to a prefactor) with the energy current $\mathbf{J}$ defined in Eq. (1.17) in chapter 1. We immediately see that by computing the flux of $\mathbf{q}$ through a surface dS, namely $\mathbf{q}(\mathbf{r},t)\cdot\mathbf{n}\,dS$, we recover Eq. (4.1).

We also define the energy density (unit J.m^{-3}), as

$$U(\mathbf{r},t) = \int_{4\pi} \frac{I(\mathbf{r},\mathbf{u},t)}{v_E}\,d\Omega \tag{4.3}$$

where v_E is the energy velocity (also known as the transport velocity).

It is interesting to examine two particular cases. For a collimated radiation propagating along a direction $\mathbf{u}_0$, the specific intensity can be written $I(\mathbf{r},\mathbf{u},t) = I_0(\mathbf{r},t)\,\delta(\mathbf{u}-\mathbf{u}_0)$, where the Dirac delta function $\delta(\mathbf{u}-\mathbf{u}_0)$ has to be understood in the sense of the angular integration over the solid angle $d\Omega$.[3] It is easy to see that in this case $\mathbf{q}(\mathbf{r},t) = v_E\mathbf{u}_0\,U(\mathbf{r},t)$. This relation takes the same form as the equation $\mathbf{j} = \rho\mathbf{v}$ that connects the current density to the charge density and the velocity of the charges in electrodynamics. This clarifies the meaning of the energy velocity introduced in Eq. (4.3). For an isotropic radiation, the specific intensity is independent on $\mathbf{u}$, and we find $I(\mathbf{r},t) = (v_E/4\pi)\,U(\mathbf{r},t)$ and $\mathbf{q}(\mathbf{r},t) = 0$.

We will now describe the evolution of the specific intensity in a scattering and absorbing medium, in order to describe the transport of the average intensity. The properties of medium will be described by averaged parameters such as the scattering and absorption mean free path.

[3] In spherical coordinates $\delta(\mathbf{u}-\mathbf{u}_0) = \delta(\theta-\theta_0)\,\delta(\phi-\phi_0)/|\sin\theta_0|$.

4.2 The RTE as an energy balance

The transport equation will result from an energy balance for the intensity propagating along the direction $\mathbf{u}$, taking the form

$$I(\mathbf{r}+d\mathbf{r},\mathbf{u},t+dt)-I(\mathbf{r},\mathbf{u},t)=-\text{losses}+\text{gain}\,, \tag{4.4}$$

where $d\mathbf{r}=ds\,\mathbf{u}$ is an infinitely small displacement along $\mathbf{u}$, and $ds=v_E\,dt$ since the energy propagates at velocity v_E.

4.2.1 Losses by scattering and absorption

We have seen in chapter 3 that the probability of scattering over a infinitely short distance ds is ds/ℓ_s, with ℓ_s the scattering mean free path. The same holds for absorption, with a probability ds/ℓ_a. Therefore the loss term in the balance equation is

$$\text{losses}=\frac{ds}{\ell_s}I(\mathbf{r},\mathbf{u},t)+\frac{ds}{\ell_a}I(\mathbf{r},\mathbf{u},t)\,. \tag{4.5}$$

This corresponds to the power taken out from direction $\mathbf{u}$ upon propagation from $\mathbf{r}$ to $\mathbf{r}+d\mathbf{r}$.

Note that one sometimes prefers to use the scattering, absorption and extinction coefficients $\mu_s=1/\ell_s$, $\mu_a=1/\ell_a$ and $\mu_e=1/\ell_e$ instead of mean free paths (this is very common in biomedical optics).

4.2.2 Gain by scattering

The scattering process redistributes the incident energy over all directions. To handle the angular distribution of scattering, we introduce the phase function $p(\mathbf{u},\mathbf{u}')$ such that $1/(4\pi)p(\mathbf{u},\mathbf{u}')$ is the fraction of power scattered from direction $\mathbf{u}'$ to direction $\mathbf{u}$. Note that the factor $1/(4\pi)$ corresponds to a normalization of the phase function such that $\int_{4\pi}p(\mathbf{u},\mathbf{u}')\,d\Omega=4\pi$.

The gain term in the energy balance results from scattering events occuring upon propagation over the distance ds, and redistributing the power towards the direction $\mathbf{u}$. Using the phase function, the gain term is

$$\text{gain}=\frac{ds}{\ell_s}\times\frac{1}{4\pi}\int_{4\pi}p(\mathbf{u},\mathbf{u}')\,I(\mathbf{r},\mathbf{u}',t)\,d\Omega'\,. \tag{4.6}$$

4.2.3 Balance equation

Using the expressions above, we find that the energy balance equation is

$$I(\mathbf{r}+d\mathbf{r},\mathbf{u},t+dt)-I(\mathbf{r},\mathbf{u},t)=-\frac{1}{\ell_e}\,I(\mathbf{r},\mathbf{u},t)ds+\frac{1}{4\pi\ell_s}\int_{4\pi}p(\mathbf{u},\mathbf{u}')\,I(\mathbf{r},\mathbf{u}',t)\,d\Omega'\,ds$$

The left-hand side can be rewritten as

$$\frac{\partial}{\partial t}I(\mathbf{r},\mathbf{u},t)\,dt+\mathbf{u}\cdot\nabla I(\mathbf{r},\mathbf{u},t)\,ds\,, \tag{4.7}$$

where the gradient operator in the second term has to be taken over the position $\mathbf{r}$. We obtain

$$\frac{1}{v_E}\frac{\partial}{\partial t}I(\mathbf{r},\mathbf{u},t)+\mathbf{u}\cdot\nabla I(\mathbf{r},\mathbf{u},t)=-\frac{1}{\ell_e}\,I(\mathbf{r},\mathbf{u},t)+\frac{1}{4\pi\ell_s}\int_{4\pi}p(\mathbf{u},\mathbf{u}')\,I(\mathbf{r},\mathbf{u}',t)\,d\Omega' \tag{4.8}$$

where we have used $dt=ds/v_E$ in the first term. This equation, known as the RTE, describes the transport of the specific intensity in a scattering and absorbing medium. It includes both partial derivatives and an integral term (it bears similarity with the Boltzmann equation used in the kinetic theory of gases). Many techniques to solve the RTE in simple geometries have been developed [1, 14, 16, 17], and analytical solutions exist only in a few particular cases (for example slab geometry with isotropic scattering). In many practical situations, we have to rely on numerical simulations. In this lecture, we will not discuss solutions to the RTE, but instead use the RTE as a step towards the diffusion approximation, which is the subject of the next chapter.

4.3 Parameters in the RTE

The parameters ℓ_s, ℓ_a, v_E and $p(\mathbf{u},\mathbf{u}')$ need to be determined for practical calculations. They correspond to average properties of the scattering medium at the frequency ω of the propagating wave.

In diluted media made of identical particles, and in the independent scattering regime (each particle scatters independently of the presence of the other particles), we have $\mu_s=1/\ell_s=\rho\,\sigma_s$, where ρ is the number density of scatterers and σ_s their scattering cross section. Likewise, for absorption we have $\mu_a=1/\ell_a=\rho\,\sigma_a$, with σ_a the absorption cross section of individual scatterers.

To characterize the relative weight of scattering and absorption in the extinction process, we can use the albedo

$$a=\frac{\mu_s}{\mu_s+\mu_a} \tag{4.9}$$

such that $a = 1$ for a purely scattering medium (*e.g.* a white paper for visible light) and $a = 0$ for a purely absorbing medium (*e.g.* concentrated black ink).

In the independent scattering regime, the phase function is simply

$$p(\mathbf{u}, \mathbf{u}') = \frac{4\pi}{\sigma_s} \frac{d\sigma_s}{d\Omega} \tag{4.10}$$

where $d\sigma_s/d\Omega$ is the differential scattering cross section of a scatterer introduced in chapter 1.

Remarks :

- In many practical situations, the medium is homogeneous and isotropic at the macroscopic scale. In this case the phase function only depends on the relative angle Θ between the incident direction $\mathbf{u}'$ and the scattering direction $\mathbf{u}$ (Θ is referred to as the scattering angle). We have $p(\mathbf{u}, \mathbf{u}') = p(\mathbf{u} \cdot \mathbf{u}') = p(\cos\Theta)$. **This condition is assumed to be satisfied throughout the rest of this lecture**.

- The degree of anisotropy of the scattering process can be measured using the anisotropy factor g defined as

$$g = \frac{1}{4\pi} \int_{4\pi} \mathbf{u} \cdot \mathbf{u}' \; p(\mathbf{u} \cdot \mathbf{u}') \, d\Omega = \frac{1}{4\pi} \int_{4\pi} \cos\Theta \; p(\cos\Theta) \, d\Omega \; . \tag{4.11}$$

  With this definition, g is the average cosine of the scattering angle (averaged over the angular distribution of scattering). $g = 0$ corresponds to isotropic scattering, and $g \simeq 1$ correspond to strong forward scattering. In a medium made of identical particles, the value of g is directly connected to the size of the particles compared to the wavelength ($g \simeq 0$ for small particles and $g \simeq 1$ for large particle).[4]

The energy velocity in its simple form equals the phase velocity c/n_{eff}, with n_{eff} the effective refractive index of the medium and c the speed of light in vacuum. The effective index is defined as the refractive index seen by the average field (see Complement B for a more precise definition of n_{eff}). This expression breaks down in the presence of sharp resonances in the scatterers, that delay the transport of the intensity. For expressions of the energy velocity in resonant multiple scattering of light, a point that will not be addressed in this lecture, see for example [1, 12].

[4] In the presence of statistical correlations in the positions of the particles, interferences between the fields scattered by different particles can lead to a phase function and a value of g that differ from that given by individual particles. In this regime the independent scattering approximation beaks down (and one speaks of dependent scattering).

4.4 Ballistic and diffuse intensities

In the situation where the scattering medium is illuminated with a collimated beam (assumed to be a plane wave) propagating along direction $\mathbf{u}_0$, we can split the specific intensity into a ballistic (or collimated) component and a diffuse component:

$$I(\mathbf{r},\mathbf{u},t) = I_{\rm b}(\mathbf{r},t)\,\delta(\mathbf{u}-\mathbf{u}_0) + I_{\rm d}(\mathbf{r},\mathbf{u},t) \tag{4.12}$$

where $\delta(\mathbf{u}-\mathbf{u}_0)$ is again the Dirac delta function with respect to the angular integration over the solid angle. Inserting this decomposition into the RTE (4.8), we obtain two equations (one for the terms involving $\delta(\mathbf{u}-\mathbf{u}_0)$, and one for the non-singular terms). For the ballistic component, we get

$$\frac{1}{v_E}\,\frac{\partial}{\partial t} I_{\rm b}(\mathbf{r},t) + \mathbf{u}_0\cdot\nabla\, I_{\rm b}(\mathbf{r},t) = -\frac{1}{\ell_e}\, I_{\rm b}(\mathbf{r},t)\ . \tag{4.13}$$

This equation leads to the result derived in section 3.3 in the steady-state regime. Indeed, choosing the Oz axis along the direction $\mathbf{u}_0$, and dropping the term $\partial/\partial t$, we find $I_{\rm b}(z) = I_{\rm b}(0)\exp(-z/\ell_e)$, which is the Beer-Lambert law. For the diffuse component, we obtain an RTE with a source term:

$$\begin{aligned}\frac{1}{v_E}\,\frac{\partial}{\partial t} I_{\rm d}(\mathbf{r},\mathbf{u},t) + \mathbf{u}\cdot\nabla\, I_{\rm d}(\mathbf{r},\mathbf{u},t) \;&=\; -\mu_e\, I_{\rm d}(\mathbf{r},\mathbf{u},t) + \frac{\mu_s}{4\pi}\int_{4\pi} p(\mathbf{u}\cdot\mathbf{u}')\, I_{\rm d}(\mathbf{r},\mathbf{u}',t)\, d\Omega' \\ &\quad + \frac{\mu_s}{4\pi}\, p(\mathbf{u}\cdot\mathbf{u}_0)\, I_{\rm b}(\mathbf{r},t)\,.\end{aligned} \tag{4.14}$$

The source term describes the transfer of energy from the ballistic beam to the diffuse intensity. The relative weight of the ballistic and diffuse components actually drives the transport regime. In particular, we will see in the next chapter that when the transport occurs through the diffuse component only, the RTE asymptotically simplifies into a diffusion equation at large length and time scales.

Chapter 5

Diffusion approximation

In this chapter, we show that at large length and time scales, the radiative transfer equation (RTE) simplifies into a diffusion equation that drives the transport of the energy density. The diffusion equation is much simpler to solve, and is a very convenient tool to analyze practical situations. Since diffusion processes are found in many transport phenomena, the diffusion equation allows one to draw interesting and useful analogies.

We have seen in chapter 4 that the specific intensity $I(\mathbf{r}, \mathbf{u}, t)$ obeys the RTE

$$\frac{1}{v_E}\frac{\partial}{\partial t} I(\mathbf{r}, \mathbf{u}, t) + \mathbf{u} \cdot \nabla\, I(\mathbf{r}, \mathbf{u}, t) = -\mu_e\, I(\mathbf{r}, \mathbf{u}, t) + \frac{\mu_s}{4\pi} \int_{4\pi} p(\mathbf{u} \cdot \mathbf{u}')\, I(\mathbf{r}, \mathbf{u}', t)\, d\Omega' \,, \quad (5.1)$$

where here we use the extinction and scattering coefficients $\mu_e = 1/\ell_e$ and $\mu_s = 1/\ell_s$. Starting from Eq. (5.1), we will show that the energy density $U(\mathbf{r}, t)$ satisfies a diffusion equation at large scales. We consider here the RTE without a source term. In the case of a scattering medium illuminated by a collimated beam, we can split the specific intensity into its ballistic and diffuse components, and the RTE for the diffuse component includes a source term, as shown in section 4.4 of the previous chapter (Eq. 4.14). For the interested reader, the derivation of the diffusion equation starting from the RTE with a source term due to an illumination by a collimated beam is given in Complement D.

5.1 Local energy conservation

The RTE can be transformed into a local conservation equation, similar to that found for the transport of particles. To proceed, we integrate Eq. (5.1) over $\mathbf{u}$ (the solid angle):

$$\frac{1}{v_E}\frac{\partial}{\partial t} \int_{4\pi} I(\mathbf{r}, \mathbf{u}, t) d\Omega + \int_{4\pi} \mathbf{u} \cdot \nabla\, I(\mathbf{r}, \mathbf{u}, t) d\Omega = -\mu_e \int_{4\pi} I(\mathbf{r}, \mathbf{u}, t) d\Omega + \mu_s \int_{4\pi} I(\mathbf{r}, \mathbf{u}', t) d\Omega' \quad (5.2)$$

where we have used the normalization of the phase function $\int_{4\pi} p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega = 4\pi$ in the last term. The definition of the energy density $U(\mathbf{r},t)$ (Eq. 4.3) directly leads to $\int_{4\pi} I(\mathbf{r},\mathbf{u},t)\,d\Omega = v_E\,U(\mathbf{r},t)$. From the definition of the energy current $\mathbf{q}(\mathbf{r},t)$ (Eq. 4.2), we have:

$$\int_{4\pi} \mathbf{u}\cdot\nabla_{\mathbf{r}}\, I(\mathbf{r},\mathbf{u},t)\,d\Omega = \nabla_{\mathbf{r}}\cdot\int_{4\pi} I(\mathbf{r},\mathbf{u},t)\,\mathbf{u}\,d\Omega = \nabla\cdot\,\mathbf{q}(\mathbf{r},t)\;.$$

Equation (5.2) can be rewritten in the form

$$\frac{\partial}{\partial t}U(\mathbf{r},t) + \nabla\cdot\,\mathbf{q}(\mathbf{r},t) + \mu_a\,v_E\,U(\mathbf{r},t) = 0\,. \tag{5.3}$$

In a non absorbing medium ($\mu_a = 0$), the equation simpifies into

$$\frac{\partial}{\partial t}U(\mathbf{r},t) + \nabla\cdot\,\mathbf{q}(\mathbf{r},t) = 0 \tag{5.4}$$

which takes the form of a local conservation equation.

5.2 First moment of the RTE

We now derive a second equation involving the energy density and the energy current. To proceed, we multiply the RTE (5.1) by $\mathbf{u}$, and we integrate over $\mathbf{u}$ (this amounts to taking the first moment of the RTE in terms of the angular variables). We obtain:

$$\begin{aligned}\frac{1}{v_E}\frac{\partial}{\partial t}\mathbf{q}(\mathbf{r},t) + \int_{4\pi}\mathbf{u}\left[\mathbf{u}\cdot\nabla\, I(\mathbf{r},\mathbf{u},t)\right]\,d\Omega \;&=\; -\mu_e\,\mathbf{q}(\mathbf{r},t) \\ &+\; \frac{\mu_s}{4\pi}\int_{4\pi}\left[\int_{4\pi}\mathbf{u}\,p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega\right] I(\mathbf{r},\mathbf{u}',t)\,d\Omega'\;.\end{aligned} \tag{5.5}$$

The integral in the right-hand side can be simplified. Using the anistropy parameter g defined in Eq. (4.11), we can show that:[1]

$$\int_{4\pi}\mathbf{u}\,p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega = 4\pi\,g\,\mathbf{u}'\;. \tag{5.6}$$

Inserting (5.6) into Eq. (5.5), we find that

$$\frac{1}{v_E}\frac{\partial}{\partial t}\mathbf{q}(\mathbf{r},t) + \left[\mu_a + \mu_s(1-g)\right]\mathbf{q}(\mathbf{r},t) = -\int_{4\pi}\mathbf{u}\,\left[\mathbf{u}\cdot\nabla\, I(\mathbf{r},\mathbf{u},t)\right]\,d\Omega\;. \tag{5.7}$$

This equation has been deduced from the RTE without any approximation. The integral term in the right-hand side remains complicated to handle, and its simplification will lead to the diffusion approximation.

[1] Take the direction of the unit vector $\mathbf{u}'$ as the (Oz) axis. The coordinates of $\mathbf{u}$ in spherical coordinates are $(\sin\theta\,\cos\phi, \sin\theta\,\sin\phi, \cos\theta)$. Due to the integration over ϕ, the only non-zero contribution of the integral $\int_{4\pi}\mathbf{u}\,p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega$ is along the (Oz) axis, and therefore along $\mathbf{u}'$. By definition of g (Eq. 4.11), this contribition is $4\pi g$. We deduce that $\int_{4\pi}\mathbf{u}\,p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega = 4\pi g\mathbf{u}'$.

5.3 Transport mean free path

In the left-hand side in Eq. (5.7) a factor $\mu_s(1-g)$ has appeared. This term actually defines a new length scale

$$\ell_t = \frac{1}{\mu_s(1-g)} = \frac{\ell_s}{1-g} \tag{5.8}$$

known as the transport mean free path. We will see that it plays an important role in the regime of diffusive transport.

In a medium with isotropic scattering ($g = 0$), we have $\ell_t = \ell_s$. Conversely, in a medium with anisotropic scattering, the two length scales can be very different. For example, in biological tissues and for near infrared light, the inhomogeneities (scatterers) have sizes on the order of the wavelength, or even larger, and $g \simeq 0,9$, $\ell_s \simeq 100\,\mu$m and $\ell_t \simeq 1$ mm. The transport mean free path has to be understood as the average distance after which the angular distribution of the intensity has become quasi-isotropic.

5.4 Deep multiple scattering

In a medium with size $L \gg \ell_s$, and with weak absorption ($\ell_a \gg \ell_s$), the wave enters the regime of multiple scattering. After a propagation distance larger than the transport mean free path ℓ_t, the angular distribution of the wave intensity becomes quasi-isotropic. The specific intensity being a smooth function of the direction, we can perform a first order expansion in terms of the angular variable $\mathbf{u}$. Technically, this amounts to expanding the specific intensity on a basis of Legendre polynomials, and keeping only the first two terms, which is known as the P_1 approximation. In this approximation, the specific intensity is [16]

$$I(\mathbf{r},\mathbf{u},t) = I^0(\mathbf{r},t) + \frac{3}{4\pi}\,\mathbf{q}(\mathbf{r},t)\cdot\mathbf{u} \tag{5.9}$$

where I^0 is an isotropic contribution and the second term is the first correction to isotropy.

We note that the integral of the anisotropic term over directions vanishes:

$$\int_{4\pi} \mathbf{q}(\mathbf{r},t)\cdot\mathbf{u}\,d\Omega = \mathbf{q}(\mathbf{r},t)\cdot\int_{4\pi}\mathbf{u}\,d\Omega = 0\ .$$

Integrating Eq. (5.9) over $\mathbf{u}$ leads to $I^0(\mathbf{r},t) = v_E\,U(\mathbf{r},t)/(4\pi)$, showing that the isotropic term in the specific intensity is proportionnal to the energy density.

5.4.1 Energy current

We can now simplify the term in the right-hand side in Eq. (5.7). Let us rewrite this term using tensor notations.[2] Its component along direction j reads:

$$\int_{4\pi} u_j \, u_i \, \frac{\partial}{\partial x_i} I(\mathbf{r}, \mathbf{u}, t) \, d\Omega \; .$$

Under the P_1 approximation, this expression can be simplified as follows:

$$\begin{aligned} \int_{4\pi} u_j \, u_i \, \frac{\partial}{\partial x_i} I(\mathbf{r}, \mathbf{u}, t) \, d\Omega &= \frac{\partial}{\partial x_i} \int_{4\pi} u_j \, u_i \, I(\mathbf{r}, \mathbf{u}, t) \, d\Omega \\ &= \frac{\partial}{\partial x_i} I^0(\mathbf{r}, t) \int_{4\pi} u_j \, u_i \, d\Omega + \frac{3}{4\pi} \frac{\partial}{\partial x_i} q_k(\mathbf{r}, t) \int_{4\pi} u_j \, u_i \, u_k \, d\Omega \; . \end{aligned}$$

It can be shown[3] that $\int_{4\pi} u_j \, u_i \, d\Omega = (4\pi/3) \, \delta_{ij}$, and $\int_{4\pi} u_j \, u_i \, u_k \, d\Omega = 0$. The component along direction j of Eq. (5.7) simplifies into

$$\frac{1}{v_E} \frac{\partial}{\partial t} q_j(\mathbf{r}, t) + \mu_s (1 - g) \, q_j(\mathbf{r}, t) = -\frac{4\pi}{3} \frac{\partial}{\partial x_j} I^0(\mathbf{r}, t) \tag{5.10}$$

where we have assumed $\mu_a \ll \mu_s(1-g)$, or equivalently $\ell_a \gg \ell_t$, and neglected the absorption term. Using the connection between U and I^0, and coming back to vector notations, we end up with:

$$\frac{1}{v_E} \frac{\partial}{\partial t} \mathbf{q}(\mathbf{r}, t) + \mu_s (1 - g) \, \mathbf{q}(\mathbf{r}, t) = -\frac{v_E}{3} \nabla U(\mathbf{r}, t) \; . \tag{5.11}$$

At sufficiently large time scales, the first term becomes negligible. Denoting by τ the characteristic time scale of the energy flux, and q the order of magnitude of the energy current, we have $1/v_E \, |(\partial/\partial t)\mathbf{q}| \sim q/(v_E \, \tau)$ and $\mu_s(1-g) \, |\mathbf{q}| \sim \mu_s(1-g) \, q = q/\ell_t$. The first term in Eq. (5.11) is negligible provided that $\ell_t \ll v_E \, \tau$. To get an order of magnitude, consider biomedical optics using near infrared light. We have $\ell_t \simeq 1$ mm, and the condition holds as long as $\tau > 0.1$ ns, which is not a severe condition in practice. Finally, the energy current and the energy density are related by

$$\mathbf{q}(\mathbf{r}, t) = -\frac{1}{3} v_E \, \ell_t \, \nabla U(\mathbf{r}, t) = -D \, \nabla U(\mathbf{r}, t) \; , \tag{5.12}$$

which takes the form of a diffusion law (similar to Fourier's law, Fick's law or Ohm's law) with a diffusion constant $D = (1/3) \, v_E \, \ell_t$. Note that this expression of the diffusion constant is typical of random walks with velocity v_E and mean free path ℓ_t in three dimensions.

[2] In these notations we denote by x_j, with $j = 1, 2, 3$, the space coordinates (instead of x, y, z), and by u_j the component of $\mathbf{u}$ along direction j. A summation is implicit each time a repeated index appears. For example, $a_i b_i$ has to be understood as $\sum_i a_i b_i$.

[3] One can use spherical coordinates. This is left as an exercise.

5.4.2 Diffusion equation

Inserting Eq. (5.12) into Eq. (5.3), we find that the energy density obeys the diffusion equation

$$\frac{\partial}{\partial t}U(\mathbf{r},t) - D\,\nabla^2 U(\mathbf{r},t) + \mu_a\, v_E\, U(\mathbf{r},t) = 0\ . \tag{5.13}$$

The last term acounts for absorption losses, and vanishes in a non absorbing medium (in this case the equation takes the form of the heat equation).

In summary, we have shown that in the deep multiple scattering regime, at large length and time scales compared to ℓ_t and ℓ_t/v_E, the energy density of the wave obeys a diffusion equation. Since this equation is far easier to handle than the RTE, it is used in many applications in imaging and sensing in (or through) scattering media. This result also shows that in the diffusive regime the transport of the average intensity of the wave can be seen as a transport of classical particles following a random walk with velocity v_E and mean free path ℓ_t.

5.5 An example of diffusive behavior

Consider a slab with thickness L, filled with a non absorbing scattering material, and illuminated by a monochromatic plane wave at normal incidence. Assuming $L \gg \ell_t$, the diffusion approximation can be used to evaluate the transmitted diffuse intensity.

Taking the (Oz) direction normal to the slab interfaces, with $z = 0$ and $z = L$ the input and output interfaces, respectively, the energy density $U(z)$ satisfies

$$\frac{d^2U}{dz^2} = 0\ .$$

After integration we obain

$$\frac{dU}{dz} = \text{constant} = \frac{-q}{D}$$

with q the z-component of the energy current (that gives the flux per unit surface flowing through the medium). A second integration leads to

$$U(z=L) - U(z=0) = \frac{-qL}{D}$$

which allows us to express the flux $\Phi = qS$ (unit W) flowing through a cross section S of the slab:

$$\Phi = \frac{D\,S}{L}\left[U(z=0) - U(z=L)\right]\ .$$

By analogy with Ohm's law, we can define the conductance G as

$$G = \frac{D\,S}{L}\,.$$

We see that the conductance scales as $1/L$, which is a feature of diffusive transport (remember that Ohm's law leads to a resistance scaling as L, which means a conductance scaling as $1/L$). We can also define a diffuse transmission coefficient T, proportionnal to G. Its calculation requires the determination of $U(z=0)$ and $U(z=L)$, taking into account the boundary conditions at the slab interfaces under illumination by a plance wave, and is performed in Complement D. The scaling of the diffuse transmission coefficient is

$$T \sim \frac{\ell_t}{L}\,.$$

This scaling explains the high reflectivity $R = 1-T$ of strongly scattering materials. Indeed, a non absorbing and scattering medium illuminated with white light, and such that $L \gg \ell_t$ for all wavelengths, strongly reflects over the full spectrum with a quasi-isotropic angular distribution. The medium appears bright and white, as a glass of milk, a sheet of paper, a thick cloud seen from a plane, or snow under sunlight.

5.6 Boundary condition for the diffusion equation

Writing a boundary condition for the diffusion equation at an interface between a scattering medium and a non scattering medium (as vacuum or air in optics) requires some care. We assume a flat interface and index matched media (the simplest situation). The geometry is represented in Fig. 5.1. Within the scattering medium (occupying the half-space $z>0$), we assume that the wave energy density obeys the steady-state diffusion equation

$$D\,\nabla^2 U(\mathbf{r},t) - \mu_a\,v_E\,U(\mathbf{r},t) = 0\,. \tag{5.14}$$

In order to derive a boundary condition at the interface $z=0$ in terms of the energy density, we use the fact that for index matched media (no internal reflection) the diffuse flux propagating towards $z>0$ vanishes at the interface [16]. In terms of the diffuse specific intensity, this condition is

$$\int_{2\pi} I_{\rm d}(z,\mathbf{u})\,\mathbf{u}\cdot\mathbf{e}_z\,d\Omega = 0 \quad \text{for} \quad z=0\,, \tag{5.15}$$

where $\mathbf{e}_z$ is the unit vector of the z-axis and the angular integration is over directions such that $\mathbf{u}\cdot\mathbf{e}_z > 0$. Under the P_1 approximation, we have (Eq. 5.9):

$$I_{\rm d}(z,\mathbf{u}) = \frac{v_E\,U(z)}{4\pi} + \frac{3}{4\pi}\,\mathbf{q}(z)\cdot\mathbf{u}\,. \tag{5.16}$$

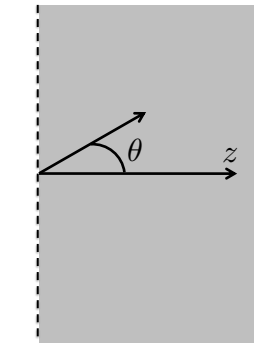

Figure 5.1: Geometry used to study the boundary conditions in the diffusion approximation. The medium $z < 0$ is non scattering, while the medium $z > 0$ is assumed to be strongly scattering, with a refractive index matched to the external medium.

Inserting this expansion into Eq. (5.15) leads to

$$\frac{v_E\, U(z=0)}{4\pi} \int_{2\pi} \mathbf{u}\cdot\mathbf{e}_z\, d\Omega + \frac{3}{4\pi}\int_{2\pi} \mathbf{q}(z=0)\cdot\mathbf{u}\,(\mathbf{u}\cdot\mathbf{e}_z) d\Omega = 0\,. \tag{5.17}$$

Defining $\cos\theta = \mathbf{u}\cdot\mathbf{e}_z$, we obtain

$$\frac{v_E\, U(z=0)}{4\pi} \int_{2\pi} \cos\theta\, d\Omega + \frac{3}{4\pi}\, q(z=0) \int_{2\pi} \cos^2\theta\, d\Omega = 0$$

which finally leads to

$$\frac{v_E}{2}\, U(z=0) + q(z=0) = 0\,. \tag{5.18}$$

Making use of the expression of the energy current Eq. (5.12), we obtain the boundary condition at the interface $z = 0$:

$$U(z=0) - \frac{2}{3}\ell_t \frac{dU}{dz}(z=0) = 0\,. \tag{5.19}$$

A pratical way to apply this boundary condition is to extrapolate $U(z)$ linearly outside the medium. We see that the energy density vanishes at a distance $z_0 = (2/3)\ell_t$, known as the extrapolation distance. Imposing $U(z) = 0$ for $z = -z_0$ is equivalent to imposing (5.19).

Finally, let us note that we have dealt with the simple case of index matched media. In the presence of a refractive index contrast, the boundary condition has to be modified. The extrapolation distance z_0 takes a different value, that accounts for internal reflection of the diffuse intensity at the interface. For a detailed study and the derivation of practical expressions, see for example Refs. [1, 18, 19, 13].

Part III

Speckle

Chapter 6

Intensity statistics

Scattering of a coherent wave by one realization of a disordered material (e.g., a solid powder, a sheet of paper, or the rough surface of a material) leads to a complex spatial distribution of intensity $I = \langle I \rangle + \delta I$ known as a speckle pattern. An example is shown in Fig. 6.1. The detailed analysis of a particular speckle pattern is most of the time out of reach and even useless. Nevertheless, speckle patterns can be characterized statistically. An interesting feature of speckle patterns is the existence of universal statistical properties, that do not depend on the details of the microstructure of the scattering medium. In this chapter, we study the statistical distribution of the intensity measured at one point in a speckle pattern.

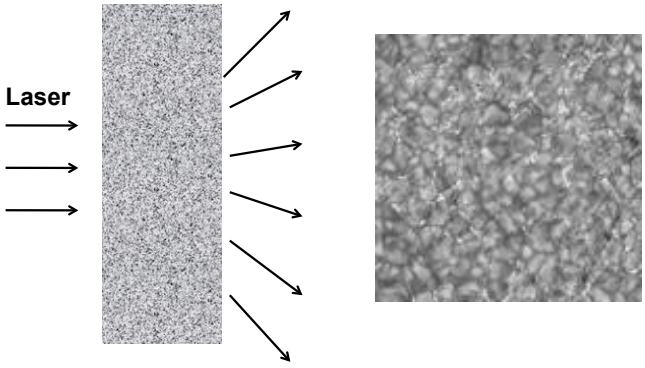

Figure 6.1: Optical speckle pattern generated by illuminating a slab of scattering medium (corresponding to one realization of disorder) with a coherent laser beam.

6.1 Field propagator and scattering sequences

The concept of scattering sequences is very useful in the study of speckles in the multiple scattering regime. Here we justify the use of this concept on the basis of theoretical elements introduced in chapter 3.

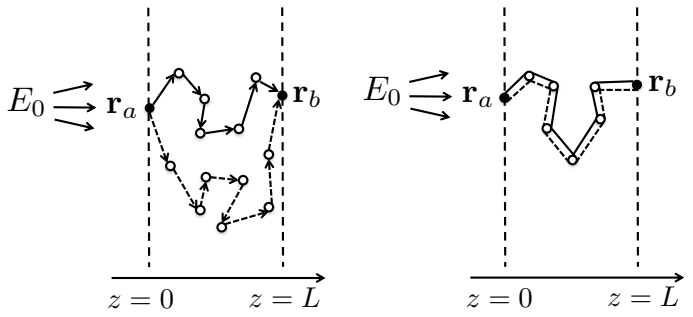

Figure 6.2: Left: Schematic representation of scattering sequences in a slab geometry for E (solid line) and E^* (dashed line). White circles stand for scattering events (scatterers). Black point are entry and exit points on the slab surfaces. Right: Leading contribution in the ladder approximation in which E and E^* follow the same scattering sequences.

Consider the canonical slab geometry shown in Fig. 6.2. The transmitted field $E(\mathbf{r}_b)$ at a point $\mathbf{r}_b$ on the output surface $z = L$ is linearly related to the incident field $E_0(\mathbf{r}_a)$ at a point $\mathbf{r}_a$ on the input surface $z = 0$. We can define a propagator $h(\mathbf{r}_b, \mathbf{r}_a)$ for the complex amplitude of the field, such that

$$E(\mathbf{r}_b) = \int_{z=0} h(\mathbf{r}_b, \mathbf{r}_a)\, E_0(\mathbf{r}_a)\, d^2\rho_a \tag{6.1}$$

where we use the notation $\mathbf{r}_a = (\boldsymbol{\rho}_a, z = 0)$ and the integral is along the input surface. We can also define a propagator for the reflected field by choosing both $\mathbf{r}_a$ and $\mathbf{r}_b$ on the input surface $z = 0$.

We will now show that the material in chapter 3 allows us to write the amplitude propagator in the form of a summation over scattering sequences

$$h(\mathbf{r}_b, \mathbf{r}_a) = \sum_{n=0}^{\infty} \sum_{\mathcal{S}_n = \{\mathbf{r}_1, \mathbf{r}_2 \ldots \mathbf{r}_n\}} A_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a)\, \exp[i\phi_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a)]\,. \tag{6.2}$$

In this expression, we define a scattering sequence $\mathcal{S}_n$ with n scattering events by the positions $\{\mathbf{r}_1, \mathbf{r}_2 \ldots \mathbf{r}_n\}$ of the successive scattering events. The summation includes all scattering sequences with n scattering events, and runs for $n = 0 \to \infty$ ($n = 0$ corresponds to free propagation from $\mathbf{r}_a$ to $\mathbf{r}_b$). We denote by $A_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a)$ the (real) amplitude resulting from

sequence $\mathcal{S}_n$ connecting $\mathbf{r}_a$ to $\mathbf{r}_b$, and $\phi_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a)$ the phase shift induced by this sequence. We often use the simplified notation

$$h(\mathbf{r}_b, \mathbf{r}_a) = \sum_{\mathcal{S}_{ab}} A_{\mathcal{S}_{ab}} \, \exp(i\phi_{\mathcal{S}_{ab}}) \tag{6.3}$$

where $\mathcal{S}_{ab}$ stands for any sequence connecting $\mathbf{r}_a$ to $\mathbf{r}_b$. Two scattering sequences are represented schematically in the left panel in Fig. 6.2.

Expression (6.2) can be deduced rigorously from Eq. (3.15). Let us write explicitly the single scattering term of Eq. (3.15) at point $\mathbf{r}_b$:

$$\sum_j \int G_0(\mathbf{r}_b, \mathbf{r}_1)\, t_j(\mathbf{r}_1, \mathbf{r}_2)\, E_0(\mathbf{r}_2) d^3r_1 d^3r_2 =$$

$$\int \left[\sum_j \int G_0(\mathbf{r}_b, \mathbf{r}_1)\, t_j(\mathbf{r}_1, \mathbf{r}_2)\, h_0(\mathbf{r}_2, \mathbf{r}_a) d^3r_1 d^3r_2 \right] E_0(\mathbf{r}_a) d^2\rho_a \, . \tag{6.4}$$

In the last line, we have used the free-space amplitude propagator $h_0(\mathbf{r}_2, \mathbf{r}_a)$ that connects the entry point $\mathbf{r}_a$ to the first scattering event.[1] All multiple scattering terms in Eq. (3.15) can be written in the same way. By comparing to Eq. (6.1), we find that

$$\begin{aligned} h &= h_0 + \sum_j G_0\, t_j\, h_0 + \sum_{j,k} G_0\, t_k\, G_0\, t_j\, h_0 \\ &+ \sum_{j\neq k, k\neq l} G_0\, t_l\, G_0\, t_k\, G_0\, t_j\, h_0 + ... \end{aligned} \tag{6.5}$$

In this compact operator notation, the integrals are implicit, but we have to keep in mind that G_0 on the left in each summation connects the last scattering event to the exit point $\mathbf{r}_b$, and that h_0 on the right connects the entry point $\mathbf{r}_a$ to the first scattering event. Expression (6.5) is exactly of the form given by Eq. (6.2), which justifies the summation over scattering sequences that is often used intuitively.

We will often have to compute second-order quantities (such as the average intensity). The most general object is the field correlation function

$$\langle E(\mathbf{r}_b) E^*(\mathbf{r}'_b) \rangle = \int_{z=0} \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}'_b, \mathbf{r}'_a) \rangle \, E_0(\mathbf{r}_a) E_0^*(\mathbf{r}'_a) \, d^2\rho_a d^2\rho'_a \, . \tag{6.6}$$

Computing $\langle E(\mathbf{r}_b) E^*(\mathbf{r}'_b) \rangle$ amounts to averaging the product of pairs of scattering sequences as that represented in the left panel in Fig. 6.2. There is no hope to be able to perform this average in the most general situation. When $\ell_s \gg \lambda$, we have seen in section 3.5 that

[1] This propagator is similar to the free-space Green function $G_0(\mathbf{r}, \mathbf{r}')$, but it connects the field at point $\mathbf{r}$ to the field at point $\mathbf{r}'$, while the Green function connects a field to a source.

the ladder approximation can be used. In this setting only the contributions resulting from two fields E and E^* following the same scattering sequences contribute to $\langle E(\mathbf{r}_b)E^*(\mathbf{r}'_b)\rangle$ as represented in the right panel in Fig. 6.2. The ladder approximation allows us to write

$$\langle h(\mathbf{r}_b,\mathbf{r}_a)h^*(\mathbf{r}'_b,\mathbf{r}'_a)\rangle \simeq P(\mathbf{r}_b,\mathbf{r}_a)\,\delta(\mathbf{r}_b-\mathbf{r}'_b)\,\delta(\mathbf{r}_a-\mathbf{r}'_a) \tag{6.7}$$

where $P(\mathbf{r}_b,\mathbf{r}_a)$ is an intensity propagator that sums the contributions of all ladder scattering sequences connecting $\mathbf{r}_a$ to $\mathbf{r}_b$. This propagator can be calculated using a transport equation for the average intensity, such as the radiative transfer equation or the diffusion approximation.

6.2 Fully developed speckle

We now turn to the study of the statistics of the intensity $I(\mathbf{r})$ measured at a point $\mathbf{r}$ in a speckle pattern. We describe the scattering medium using an ensemble of realizations of disorder, and $I(\mathbf{r})$ is a random variable. Assuming ergodicity, the statistical properties of $I(\mathbf{r})$ can be compared to that obtained in practice from the spatial fluctuations of the intensity on a given image of a speckle pattern, as that shown in Fig. 6.1.

The field at a point $\mathbf{r}$ in the speckle pattern is of the form

$$E(\mathbf{r},t) = \mathrm{Re}[E(\mathbf{r})\,\exp(-i\omega t)] \tag{6.8}$$

where $\omega = 2\pi c/\lambda$ is the frequency of the incident light, λ being the wavelength in vacuum. This field is the superposition of scattered waves emerging from all possible scattering sequences inside the medium. Its complex amplitude can be written

$$E(\mathbf{r}) = \sum_{\mathcal{S}} A_{\mathcal{S}}(\mathbf{r})\,\exp[i\phi_{\mathcal{S}}(\mathbf{r})] \tag{6.9}$$

where $\mathcal{S}$ denotes any scattering sequence that starts on the entry surface of the medium and ends up at the observation point $\mathbf{r}$. Each term in the sum involves a real amplitude $A_{\mathcal{S}}(\mathbf{r})$ and a phase $\phi_{\mathcal{S}}(\mathbf{r})$ that are both random variables. Sums as in Eq. (6.9) are usually denoted by *random phasor sums* [20, 21].

The statistical properties of the intensity produced by a field of the form (6.9) can be obtained using the model of fully developed speckle, that relies on the following assumptions:

1. For two different sequences $\mathcal{S}$ and $\mathcal{S}'$, the complex amplitudes $E_{\mathcal{S}}(\mathbf{r}) = A_{\mathcal{S}}(\mathbf{r})\,\exp[i\phi_{\mathcal{S}}(\mathbf{r})]$ and $E_{\mathcal{S}'}(\mathbf{r}) = A_{\mathcal{S}'}(\mathbf{r})\,\exp[i\phi_{\mathcal{S}'}(\mathbf{r})]$ are independent random variables;
2. For a given sequence $\mathcal{S}$, the amplitude $A_{\mathcal{S}}(\mathbf{r})$ and the phase $\phi_{\mathcal{S}}(\mathbf{r})$ are mutually uncorrelated;
3. The phase $\phi_{\mathcal{S}}(\mathbf{r})$ is uniformly distributed on $[-\pi,+\pi]$.

We will see that under these assumptions, the scattered field obeys Gaussian statistics.

6.3 Amplitude distribution function

The sum (6.9) contains a large number of terms, that are independent random variables. In these conditions, it is possible to deduce the statistics of the field amplitude using the central limit theorem. Let $X = \mathrm{Re}\, E(\mathbf{r})$ and $Y = \mathrm{Im}\, E(\mathbf{r})$ be the real and imaginary parts of the field, that can be written as

$$\begin{aligned} X &= \sum_{\mathcal{S}} A_{\mathcal{S}}(\mathbf{r})\, \cos\phi_{\mathcal{S}}(\mathbf{r}) \\ Y &= \sum_{\mathcal{S}} A_{\mathcal{S}}(\mathbf{r})\, \sin\phi_{\mathcal{S}}(\mathbf{r})\ . \end{aligned}$$

The average value, variance and cross-correlation of these two random variables can be easily determined using the three hypotheses of the previous section. Since the amplitudes and phases are uncorrelated, and the phases are uniformy distributed on $[-\pi, +\pi]$, one immediately obtains $\langle X\rangle = 0$ and $\langle Y\rangle = 0$. It is also possible to compute the second moment:

$$\begin{aligned} \langle X^2\rangle &= \sum_{\mathcal{S}}\sum_{\mathcal{S}'} \langle A_{\mathcal{S}}(\mathbf{r}) A_{\mathcal{S}'}(\mathbf{r})\rangle\, \langle \cos\phi_{\mathcal{S}}(\mathbf{r}) \cos\phi_{\mathcal{S}'}(\mathbf{r})\rangle \\ &= \sum_{\mathcal{S}} \langle A^2_{\mathcal{S}}(\mathbf{r})\rangle\, \langle \cos^2\phi_{\mathcal{S}}(\mathbf{r})\rangle \\ &= \frac{1}{2}\sum_{\mathcal{S}} \langle A^2_{\mathcal{S}}(\mathbf{r})\rangle \end{aligned} \tag{6.10}$$

where we have used hypotheses 1 in the second line. The same results holds for $\langle Y^2\rangle$, and we will use the notation $\sigma^2 = \langle X^2\rangle = \langle Y^2\rangle$ for the variance of X and Y. Moreover, it is also easy to verify that $\langle XY\rangle = 0$, showing that the real and imaginary parts of the field are uncorrelated.

From the central limit theorem we can infer that, in the limit of an infinite number of terms in the summation (6.9), both X and Y are Gaussian variables with zero mean and equal variance. Since X and Y are uncorrelated (and therefore independent), the joint probability of X and Y is also Gaussian:

$$p(X,Y) = \frac{1}{2\pi\sigma^2} \exp\left(-\frac{X^2+Y^2}{2\sigma^2}\right)\ . \tag{6.11}$$

The statistics of the field amplitude $A = \sqrt{X^2+Y^2}$ is obtained from a simple change of variable:

$$\begin{aligned} p(A) &= \frac{A}{\sigma^2} \exp\left(-\frac{A^2}{2\sigma^2}\right) \quad \text{for} \quad A > 0 \\ p(A) &= 0 \quad \text{for} \quad A < 0\ . \end{aligned} \tag{6.12}$$

6.4 Intensity distribution function

The probability density of the intensity $I(\mathbf{r}) = |E(\mathbf{r})|^2$ directly follows from Eq. (6.12), using the relationship

$$p(I) = p(A = \sqrt{I})\,\frac{dA}{dI} = \frac{1}{2\sqrt{I}}\,p(A = \sqrt{I})\ . \tag{6.13}$$

We finally obtain:

$$\begin{aligned} p(I) &= \frac{1}{\langle I\rangle}\exp\left(-\frac{I}{\langle I\rangle}\right) \quad \text{for} \quad I > 0 \\ p(I) &= 0 \quad \text{for} \quad I < 0\ . \end{aligned} \tag{6.14}$$

In this expression $\langle I\rangle = 2\sigma^2$ is the averaged intensity of the speckle pattern (remember that $\langle E\rangle = 0$ in this model so that $\langle I\rangle = \langle |E|^2\rangle = \langle X^2 + Y^2\rangle = 2\sigma^2$). This form of the statistical distribution of the intensity is known as the Rayleigh statistics, and is a feature of speckle patterns in the Gaussian approximation. It is interesting to note that the most likely value of the intensity is $I = 0$.

6.5 Speckle contrast

In order to characterize the intensity fluctuations in a speckle pattern, we can calculate the variance $\mathrm{Var}(I) = \langle I^2\rangle - \langle I\rangle^2$. The speckle contrast is defined as the normalized standard deviation $\sigma_I/\langle I\rangle$ with $\sigma_I = \sqrt{\mathrm{Var}(I)}$. The second moment of the intensity is readily obtained from the probability density using an integration by part:

$$\langle I^2\rangle = \int_0^\infty I^2\,p(I)\,dI = 2\,\langle I\rangle^2\ . \tag{6.15}$$

We end up with

$$\mathrm{Var}(I) = \langle I\rangle^2 \tag{6.16}$$

which is a feature of the Rayleigh statistics. In terms of speckle contrast, this is equivalent to

$$\frac{\sigma_I}{\langle I\rangle} = 1\ . \tag{6.17}$$

A speckle pattern exhibits a large contrast, with intensity fluctuations on the same order as the averaged value. This is consistent with the fact that the intensity very frequently drops to zero.

6.6 Intensity statistics of unpolarized light

Expression (6.14) describes the intensity statistics for a scalar wave. In optics, this would correspond to the intensity of a linearly polarized field, measured after a polarizer. Finding the statistical distribution of the full intensity, measured without reducing the field to one of its components, is a more complicated task. Here we show how to find the statistics in the particular case of a speckle produced with unpolarized light.

When the speckle pattern is observed in the far field, the field is locally equivalent to a plane wave and only two components E_α and E_β need to be accounted for. The statistical distribution of the intensity $I = |E_\alpha|^2 + |E_\beta|^2$ can be deduced from that of $I_\alpha = |E_\alpha|^2$ and $I_\beta = |E_\beta|^2$. Since for unpolarized light the full intensity I is the sum of the two independent random variables I_α and I_β, its probability density $p(I)$ is the convolution product of the probability densities of I_α and I_β, both being given by Eq. (6.14). Noticing that $\langle I_\alpha \rangle = \langle I_\beta \rangle = \langle I \rangle / 2$, the convolution product takes the form:

$$p(I) = \left(\frac{2}{\langle I \rangle} \right)^2 \int_0^I \exp\left[-\frac{2(I-x)}{\langle I \rangle} \right] \exp\left(-\frac{2x}{\langle I \rangle} \right) dx \tag{6.18}$$

and $p(I) = 0$ for $I < 0$. This leads immediately to the final result

$$\begin{aligned} p(I) &= \left(\frac{2}{\langle I \rangle} \right)^2 I \exp\left(-\frac{2I}{\langle I \rangle} \right) \quad \text{for} \quad I > 0 \\ p(I) &= 0 \quad \text{for} \quad I < 0 \ . \end{aligned} \tag{6.19}$$

In this modified Rayleigh statistics, that applies to a two-dimensional unpolarized field, the most likely value of the intensity is not zero. Moreover, this distribution leads to a reduced speckle contrast $\sigma_I / \langle I \rangle = 1/\sqrt{2}$ (the calculation of the speckle contrast for unpolarized light is left as an exercise).

Chapter 7

Dynamic light scattering

In this chapter we study light scattering by an ensemble of particles in motion. A typical system is a colloidal suspension with particles under Brownian motion. Another example is a biological tissue in the presence of blood flow. In this situation, the scattered intensity fluctuates in time, as the result of the time dependent phase shifts between the fields scattered by different particles. We will show that the time fluctuations of the field or the intensity carry information on the dynamics of the particles. For the sake of illustration, we will focus on Brownian motion of particles in a fluid. Dynamic light scattering is a widespread technique in soft matter physics and in biomedical optics.

7.1 Single scattering regime

A typical geometry in a dynamic light scattering experiment is sketched in Fig. 7.1. The figure represents a configuration using transmitted light, but the analysis developed in this chapter is valid for both reflection and transmission geometries.

In the single scattering regime, we assume that the sample size is $L \sim \ell_s$, with ℓ_s the scattering mean free path. The medium is illuminated by a monochromatic plane wave with frequency ω, complex amplitude E_0 and wavevector $\mathbf{k}_i$. One measures the field scattered in a direction defined by wavevector $\mathbf{k}_s$. This field results from the superposition of the waves scattered by all scatterers. Due to their motion, the phase shifts between the scattered waves change in time, and the amplitude of the scattered field fluctuates in time. We assume that the fluctuations occur on a time scale much larger than $2\pi/\omega$, so that the field remains quasi-monochromatic, and can be written in the form $E(t)\exp(-i\omega t)$, with $E(t)$ a slowly varying complex amplitude. Note that since the motion of the scatterers induces Doppler shifts that remain small compared to ω, dynamic light scattering (DLS) in the single scattering regime is also referred to as Quasi-Elastic Light Scattering (QELS) [22].

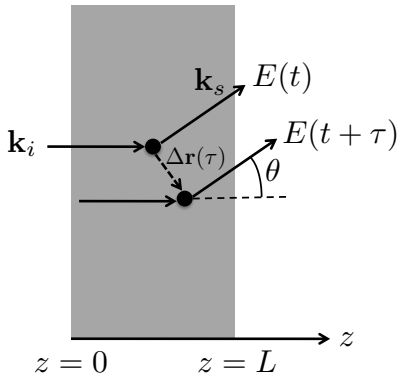

Figure 7.1: Dynamic light scattering in the single scattering regime. $\Delta\mathbf{r}(\tau)$ is the displacement of a scatterer between times t and $t+\tau$. This displacement induces a phase shift between the scattered fields $E(t)$ and $E(t+\tau)$.

In order to characterize the field fluctuations in time, we introduce the field correlation function $G_1(\tau) = \langle E(t)\, E^*(t+\tau)\rangle$, where $\langle ...\rangle$ denotes an average over the motion of the particles. In the single-scattering regime, the far-field scattered amplitude in the direction defined by wavevector $\mathbf{k}_s$ reads

$$E(t) = S(q)\,\frac{\exp(ik_0 r)}{r}\,E_0\,\sum_j \exp\left[-i\mathbf{q}\cdot\mathbf{r}_j(t)\right] \tag{7.1}$$

where $\mathbf{q} = \mathbf{k}_s - \mathbf{k}_i$, $k_0 = \omega/c = 2\pi/\lambda$, $\mathbf{r}_j(t)$ the position of particle number j at time t, and $S(q)$ the scattering amplitude of a single scatterer (introduced in chapter 1) which is assumed to depend only on $q = |\mathbf{q}|$. This leads to the following expression of the time correlation function

$$G_1(\tau) = |S(q)|^2 \frac{|E_0|^2}{r^2}\sum_j \langle \exp\left[i\mathbf{q}\cdot\Delta\mathbf{r}_j(\tau)\right]\rangle \tag{7.2}$$

where $\Delta\mathbf{r}_j(\tau) = \mathbf{r}_j(t+\tau) - \mathbf{r}_j(t)$ is the displacement of particle number j between time t and time $t+\tau$. In this expression we have also assumed that the displacements of two different particles are uncorrelated. For an ensemble of N identical particles, the average value in the summation is the same for all particles, and we end up with

$$G_1(\tau) = N\,|S(q)|^2 \frac{|E_0|^2}{r^2}\,\langle \exp\left[i\mathbf{q}\cdot\Delta\mathbf{r}(\tau)\right]\rangle\,. \tag{7.3}$$

The average in this equation has to be taken over the random variable $\Delta\mathbf{r}(\tau)$. In the case of three-dimensional Brownian motion with a diffusion constant D_B, the statistical distribution of the displacements is isotropic and Gaussian, with a probability density

$$P[\Delta r(\tau)] = \frac{1}{(4\pi\, D_B\,\tau)^{3/2}}\,\exp\left[\frac{-\Delta r(\tau)^2}{4D_B\tau}\right] \tag{7.4}$$

where $\Delta r(\tau) = |\Delta \mathbf{r}(\tau)|$. The average in Eq. (7.3) can be performed analytically. Introducing the normalized correlation function $g_1(\tau) = G_1(\tau)/G_1(0)$ for convenience, we have

$$\begin{aligned} g_1(\tau) &= \langle \exp[i\mathbf{q}\cdot\Delta\mathbf{r}(\tau)]\rangle \\ &= \langle \exp[iq_x\Delta x(\tau)]\rangle \langle \exp[iq_y\Delta y(\tau)]\rangle \langle \exp[iq_z\Delta z(\tau)]\rangle \end{aligned} \tag{7.5}$$

where each average term can be calculated using the result

$$\int_{-\infty}^{+\infty} \exp(ipX)\exp(-aX^2/2)\,dX = (2\pi/a)^{1/2}\,\exp[-p^2/(2a)] \tag{7.6}$$

and the probability density in Eq. (7.4). We end up with

$$g_1(\tau) = \exp\left(-D_B\,q^2\,\tau\right). \tag{7.7}$$

The modulus of the scattered wavevector is $q = 2\,k_0\,\sin(\theta/2)$, with θ the scattering angle defined in Fig. 7.1. Equation (7.7) shows that from a measurement of $g_1(\tau)$ one can deduce the diffusion constant D_B of the Brownian particles. A widespread application of DLS in the single scattering regime is the measurement of the size of colloidal particles. Indeed, D_B is connected to the radius R of the particles and the viscosity η of the fluid through the Einstein relation $D_B = k_B T/(6\pi\eta R)$, with T the temperature and k_B the Boltzmann constant.

7.2 Multiple scattering regime. Diffusing-Wave Spectroscopy

When the system size L becomes larger than the scattering mean free path ℓ_s, the single scattering approximation is no more valid. With reference to Fig. 7.2, we shall now study the time fluctuations of the field resulting from the superposition of multiply scattered waves.

It is convenient to use the representation of the field as a summation over scattering sequences, as described in chapter 3. The amplitude of the scattered field $E(t)$ can be written

$$E(t) = E_0 \sum_{n=1}^{\infty} \sum_{\mathcal{S}_n} A_{\mathcal{S}_n}(t)\,\exp[i\phi_{\mathcal{S}_n}(t)] \tag{7.8}$$

where E_0 is the amplitude of the incident plane wave. In this representation, a scattering sequence with n scattering events is written as $\mathcal{S}_n = \{\mathbf{r}_1(t), \mathbf{r}_2(t)...\mathbf{r}_n(t)\}$, where $\mathbf{r}_j(t)$ is the position of scatterer number j at time t. The change in amplitude and phase created by the sequence $\mathcal{S}_n$ are $A_{\mathcal{S}_n}(t)$ and $\phi_{\mathcal{S}_n}(t)$, respectively. The time correlation function of the field directly follows:

$$G_1(\tau) = |E_0|^2 \sum_{n} \sum_{\mathcal{S}_n} \sum_{n'} \sum_{\mathcal{S}'_{n'}} \langle\, A_{\mathcal{S}_n}(t) A_{\mathcal{S}'_{n'}}(t+\tau)\,\exp[i\phi_{\mathcal{S}_n}(t)]\exp[-i\phi_{\mathcal{S}'_{n'}}(t+\tau)]\,\rangle \tag{7.9}$$

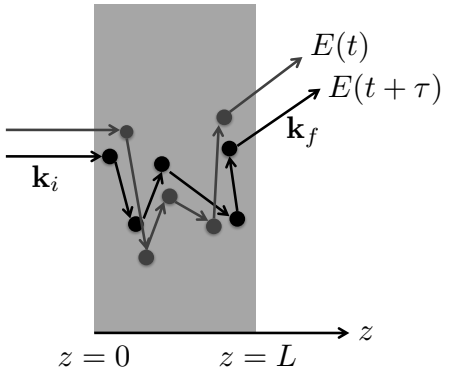

Figure 7.2: Schematic representation of the field resulting from a scattering sequence with multiple scattering events, at two differnet times t and $t+\tau$. As a consequence of the motion of scatterers, the accumulated phase shifts along the sequences at t and $t+\tau$ are different,

where $\langle ... \rangle$ denotes an average over the motion of the particles. Under the assumptions of a fully developed speckle (see chapter 6), the complex amplitude resulting from two different sequences are uncorrelated, and the output amplitude and phase shift for a given sequence are also uncorrelated. The preceding expression can be simplified into

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle\, A_{\mathcal{S}_n}(t) A_{\mathcal{S}_n}(t+\tau) \,\rangle \, \langle \exp[i\phi_{\mathcal{S}_n}(t) - i\phi_{\mathcal{S}_n}(t+\tau)] \,\rangle. \tag{7.10}$$

Due to the random motion of the particles, the phase shift along a scattering sequence will decorrelate much faster than the amplitude when τ increases. We can assume $\langle A_{\mathcal{S}_n}(t) A_{\mathcal{S}_n}(t+\tau)\rangle \simeq \langle A^2_{\mathcal{S}_n} \rangle$ and write

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \, \langle \exp[i\phi_{\mathcal{S}_n}(t) - i\phi_{\mathcal{S}_n}(t+\tau)] \,\rangle \; . \tag{7.11}$$

We are left with the evaluation of the average of the phase term. The phase difference due to the motion of the particles, along a sequence with n scattering events, can be written using the scattered wavevector $\mathbf{q}_j$ of an individual scattering event, leading to

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \left\langle \exp\left[i \sum_{j=1}^{n} \mathbf{q}_j \cdot \Delta\mathbf{r}_j(\tau) \right] \right\rangle \; . \tag{7.12}$$

In the averaging process, both $\mathbf{q}_j$ and $\Delta\mathbf{r}_j(\tau)$ are random variables (this is a major difference with the single scattering regime where $\mathbf{q}$ was fixed). An exact calculation would require handling the correlation between the scattered wavevectors $\mathbf{q}_j$ and $\mathbf{q}_{j+1}$ between successive scattering events, as well as the constraint $\sum_j \mathbf{q}_j = \mathbf{k}_f - \mathbf{k}_i$, with $\mathbf{k}_i$ and $\mathbf{k}_f$ the incident and

observation wavevectors. Although this can be done in numerical simulations, a closed-form expression can be obtained only at the cost of a series of approximations. We first assume that the average over $\mathbf{q}_j$ is independent on the motion of the particles, and depends only on the phase function of an individual scatterer and on the number of scattering events. In these conditions, the averaging processes over $\mathbf{q}_j$ and $\Delta\mathbf{r}_j(\tau)$ are considered as independent. For Brownian motion, the average over $\Delta\mathbf{r}_j(\tau)$ is performed using Eq. (7.7). We obtain

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A_{\mathcal{S}_n}^2 \rangle \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2\right) \right\rangle \tag{7.13}$$

where $q_j = |\mathbf{q}_j|$. The average over q_j remains to be performed. As the result must be independent on the scattering sequence, it is useful to introduce

$$P(n) = \sum_{\mathcal{S}_n} \langle A_{\mathcal{S}_n}^2 \rangle \tag{7.14}$$

as the fraction of the incident power that has undergone n scattering events, and rewrite Eq. (7.13) in the form

$$G_1(\tau) = |E_0|^2 \sum_n P(n) \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2\right) \right\rangle . \tag{7.15}$$

To evaluate the average over q_j^2, we use a first-cumulant expansion:

$$\begin{aligned} \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2\right) \right\rangle &\simeq \left\langle 1 - D_B\,\tau \sum_{j=1}^{n} q_j^2 \right\rangle \\ &= 1 - D_B\,\tau \sum_{j=1}^{n} \langle q_j^2 \rangle \\ &= 1 - n\,D_B\,\tau \langle q_j^2 \rangle \\ &\simeq \exp(-n\,D_B\,\tau \langle q_j^2 \rangle) . \end{aligned} \tag{7.16}$$

If θ denotes the scattering angle of an elementary scattering process, one has

$$\langle q_j^2 \rangle = 2k_0^2 \langle 1 - \cos\theta \rangle = 2k_0^2 (1 - g) = 2k_0^2 \frac{\ell_s}{\ell_t} \tag{7.17}$$

where g is the anisotropy factor and $\ell_t = \ell_s/(1-g)$ is the transport mean free path. With these simplifications, the field correlation function finally reads

$$G_1(\tau) = |E_0|^2 \sum_n P(n) \exp\left(-2k_0^2 \frac{\ell_s}{\ell_t}\, n\, D_B\, \tau\right). \tag{7.18}$$

For practical calculations, it is often easier to manipulate an integral instead of a series. To proceed, we introduce the length s of a scattering sequence with n scattering events, through $s = n\ell_s$ (note that this relation is in principle only valid on average). Using the probability density $P(s)$ of a sequence with length s, we get the final expression of the normalized field correlation function:

$$g_1(\tau) = \int_0^\infty P(s)\, \exp\left(-2\frac{\tau}{\tau_0}\,\frac{s}{\ell_t}\right)\, ds \tag{7.19}$$

with $\tau_0 = (k_0^2\, D_B)^{-1}$. This expression is widely used in the analysis of dynamic light scattering experiments in the multiple scattering regime. The associated technique is often referred to as Diffusing Wave Spectroscopy (DWS) [23].

The probability density $P(s)$, also called path-length distribution, can be obtained as the solution of a time-dependent transport equation, as the radiative transfer equation (RTE) or the diffusion equation. Indeed, from the time-dependent output flux $\phi(t)$ resulting from an incident pulse $I_{inc}\,\delta(t)$, one deduces $P(s) = \phi(t = s/v_E)/I_{inc}$ where v_E is the energy velocity in the medium (assumed to be uniform). Moreover, since Eq. (7.19) is mathematically a Laplace transform, on gets $g_1(\tau)$ directly from the solution of the RTE or diffusion equation in the Laplace domain [23].

Example

Consider a thick slab (assumed to be semi-infinite), illuminated by a plane wave, and collection of light at one point in the surface. The explicit calculation of the integral in Eq. (7.19) can be performed analytically (not shown) [23]. The result is easily expressed in terms of the variable $x = \sqrt{6\tau/\tau_0}$. A short time scale $x \ll 1$, it is simply

$$g_1(\tau) \simeq \exp(-\gamma x) \tag{7.20}$$

with $\gamma = 5/3$ (this factor results from the boundary condition in the diffusion approximation). By fitting the exponential decay of the correlation function at short time, one can deduce τ_0 and D_B.

7.3 Measured signal. Siegert relation

In practice one often measures the intensity correlation function $G_2(\tau) = \langle I(t)\, I(t+\tau)\rangle$, with $I(t) = |E(t)|^2$, instead of the field correlation $G_1(\tau)$. For fully developed speckles, a simple relation exists between $G_2(\tau)$ and $G_1(\tau)$.

For Gaussian random variables, high-order correlation functions factorize as products of second-order correlation functions. Keeping only the non-vanishing terms in the factorization, we find

that

$$\begin{aligned} G_2(\tau) &= \langle E(t)E^*(t)E(t+\tau)E^*(t+\tau)\rangle \\ &= \langle E(t)E^*(t)\rangle\langle E(t+\tau)E^*(t+\tau)\rangle + \langle E(t)E^*(t+\tau)\rangle\langle E^*(t)E(t+\tau)\rangle \end{aligned} \quad (7.21)$$

Since the statistics is assumed to be stationnary in time, the first term in the right-hand side is $G_1^2(0) = \langle I(t)\rangle^2$. The second term is $|G_2(\tau)|^2$. The relation takes the simple form

$$G_2(\tau) = \langle I(t)\rangle^2 + |G_1(\tau)|^2. \quad (7.22)$$

In practice it is useful to introduce normalized correlation functions for the field and the intensity, defined as $g_1(\tau) = \langle E(t)\, E^*(t+\tau)\rangle / \langle |E(t)|^2\rangle$ and $g_2(\tau) = \langle I(t)\, I(t+\tau)\rangle / \langle I(t)\rangle^2$. In terms of normalized correlation functions, the relation simplifies into

$$g_2(\tau) = 1 + |g_1(\tau)|^2\,, \quad (7.23)$$

which is known as the Siegert relation. It shows that the intensity correlation function can be obtained from the square modulus of the field correlation function. This relation is frequently used in the analysis of DLS experiments. Note that taken at $\tau = 0$, the relation (7.22) leads to $\langle I^2\rangle = 2\langle I\rangle^2$, which is a feature of the Rayleigh statistics derived in chapter 6.

Chapter 8

Coherent backscattering

The measurement of the angular dependence of the light intensity reflected from a thick scattering medium reveals the existence of a peak in the exact backscattering direction. This effect, known as coherent backscattering, is a signature of the underlying coherence of the multiple scattering process, and a consequence of the reciprocity theorem in wave physics. Coherent backscattering is an example of a mesoscopic phenomenon, in which the wave nature of the transport process plays a crucial role.

8.1 Reflected far field

We consider a thick scattering medium (assumed semi-infinite) illuminated by a plane wave with wavevector $\mathbf{k}_a$, as represented in Fig. 8.1. The reflected intensity is observed in the far field in a direction defined by wavevector $\mathbf{k}_b$. We define the direction Oz as being perpendicular to the interface (the latter coincinding with the plane $z = 0$), and use specific notations for the projection of vectors along the (Ox, Oy) plane, such that $\mathbf{r}_a = (\boldsymbol{\rho}_a, z = 0)$, $\mathbf{k}_a = [\mathbf{q}_a, k_z(q_a)]$, etc.

The scattered field in the plane $z = 0$ is linearly related to the incident field $E_0 \exp(i\mathbf{k}_a \cdot \mathbf{r})$. Using the amplitude propagator $h(\mathbf{r}_b, \mathbf{r}_a)$ introduced in chapter 3, it can be written as

$$E(\mathbf{r}_b) = \int_{z=0} h(\mathbf{r}_b, \mathbf{r}_a)\, E_0\, \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a)\, d^2\rho_a \tag{8.1}$$

where $\mathbf{r}_a = (\boldsymbol{\rho}_a, z = 0)$ and $\mathbf{r}_b = (\boldsymbol{\rho}_b, z = 0)$ are the input and output points. The field scattered in direction $\mathbf{k}_b$ in the far field is of the form

$$E(\mathbf{r}) = \frac{k_z(q_b)}{2i\pi}\, E(\mathbf{q}_b)\, \frac{\exp(ik_0 r)}{r} \tag{8.2}$$

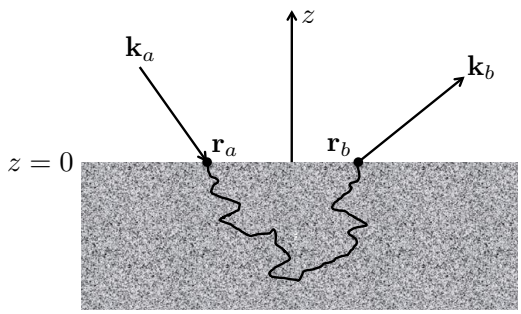

Figure 8.1: Reflection on a scattering medium illuminated by an incident plane wave with wavevector $\mathbf{k}_a$. One observes the far-field intensity reflected along direction $\mathbf{k}_b$.

where $k_z(q_b) = (k_0^2 - \mathbf{q}_b^2)^{1/2}$ is the component of $\mathbf{k}_b$ along the z direction, $k_0 = \omega/c = 2\pi/\lambda$, and $E(\mathbf{q}_b)$ is the Fourier transform of the scattered field in the plane $z = 0$:

$$E(\mathbf{q}_b) = \int_{z=0} E(\boldsymbol{\rho}_b, z = 0)\, \exp(-i\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_b \; . \tag{8.3}$$

Equation (8.2) can be obtained using the plane wave expansion of the scattered field, and taking the asymptotic expression when $k_0 r \to \infty$ (see for example [24]). Denoting by $E(\mathbf{k}_a, \mathbf{k}_b)$ the reflected amplitude in direction $\mathbf{k}_b$ in the far field, that we define as $E(\mathbf{k}_a, \mathbf{k}_b) = q_b E(\mathbf{K}_b)$ (we forget the factor $1/(2i\pi)$), we have

$$\begin{aligned} E(\mathbf{k}_a, \mathbf{k}_b) &= k_z(q_b) \int_{z=0} E(\boldsymbol{\rho}_b, z = 0)\, \exp(-i\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_b \\ &= k_z(q_b)\, E_0 \int_{z=0} h(\mathbf{r}_b, \mathbf{r}_a)\, \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a - i\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_a\, d^2\rho_b \; . \end{aligned} \tag{8.4}$$

8.2 Reflected diffuse intensity

The reflected diffuse intensity in direction $\mathbf{k}_b$ is $I_d(\mathbf{k}_a, \mathbf{k}_b) = \langle |E(\mathbf{k}_a, \mathbf{k}_b)|^2 \rangle$. Using Eq. (8.4) it takes the following form:

$$\begin{aligned} I_d(\mathbf{k}_a, \mathbf{k}_b) &= k_z^2(q_b)\, |E_0|^2 \int \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'}) \rangle\, \exp[i\mathbf{q}_a \cdot (\boldsymbol{\rho}_a - \boldsymbol{\rho}_{a'})] \\ &\quad \times \exp[-i\mathbf{q}_b \cdot (\boldsymbol{\rho}_b - \boldsymbol{\rho}_{b'})]\, d^2\rho_a d^2\rho_b d^2\rho_{a'} d^2\rho_{b'}. \end{aligned} \tag{8.5}$$

In the limit $k_0 \ell_s \gg 1$, the ladder approximation is expected to give the leading contribution, corresponding to the diagrammatic representation in Fig. 8.2.

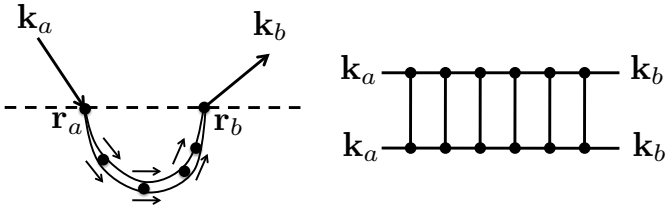

Figure 8.2: Graphical (left) and diagrammatic (right) representations of the diffuse reflection process in the ladder approximation.

As discussed in section 6.1, in the ladder approximation we can write the correlator of the amplitude propagator as (see Eq. 6.7)

$$\langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'}) \rangle \simeq P(\mathbf{r}_b, \mathbf{r}_a)\, \delta(\mathbf{r}_a - \mathbf{r}_{a'}) \delta(\mathbf{r}_b - \mathbf{r}_{b'}) \tag{8.6}$$

where $P(\mathbf{r}_b, \mathbf{r}_a)$ is the intensity propagator connecting $\mathbf{r}_a$ to $\mathbf{r}_b$. In the reflection geometry considered here, and assuming statistical translational invariance of the medium along the $x - y$ plane, it only depends on $\boldsymbol{\rho}_b - \boldsymbol{\rho}_a$. We finally end up with

$$I_d(\mathbf{k}_a, \mathbf{k}_b) = k_z^2(q_b)\, |E_0|^2 \int P(\boldsymbol{\rho}_b - \boldsymbol{\rho}_a)\, d^2\rho_a d^2\rho_b \; . \tag{8.7}$$

8.3 Reciprocity of the amplitude propagator

The amplitude propagator is connected to the reflection part of the scattering matrix $r(\mathbf{q}, \mathbf{q}')$. Indeed, by definition of the scattering matrix, the Fourier transform of the reflected field in the plane $z = 0$, defined as

$$E_s(\mathbf{q}) = \int E(\boldsymbol{\rho}) \exp(-i\mathbf{q} \cdot \boldsymbol{\rho})\, d^2\rho \; , \tag{8.8}$$

is connected to the Fourier transform of the incident field by

$$E(\mathbf{q}) = \int r(\mathbf{q}, \mathbf{q}')\, E_{inc}(\mathbf{q}')\, d^2q'. \tag{8.9}$$

From Eqs. (8.1) and (8.9) it is easy to show that

$$h(\mathbf{r}, \mathbf{r}') = \int r(\mathbf{q}, \mathbf{q}') \, \exp(i\mathbf{q} \cdot \boldsymbol{\rho} - i\mathbf{q}' \cdot \boldsymbol{\rho}') \, \frac{d^2q \, d^2q'}{4\pi^2} \tag{8.10}$$

where $\mathbf{r} = (\boldsymbol{\rho}, z = 0)$ and $\mathbf{r}' = (\boldsymbol{\rho}', z = 0)$ are two points on the interface. The amplitude propagator is therefore the Fourier transform of the reflection part of the scattering matrix. Inverting (8.10) yields

$$r(\mathbf{q}, \mathbf{q}') = \int h(\mathbf{r}, \mathbf{r}') \, \exp(-i\mathbf{q} \cdot \boldsymbol{\rho} + i\mathbf{q}' \cdot \boldsymbol{\rho}') \, d^2\rho \, d^2\rho'. \tag{8.11}$$

In any linear medium, and for materials with symmetric constitutive dielectric and permittivity tensors, the reciprocity theorem holds. In terms of the scattering matrix and for a reflection geometry, it reads (for derivations of the reciprocity relations, see for example Ref. [24])

$$k_z(q) \, r(\mathbf{q}, \mathbf{q}') = k_z(q') \, r(-\mathbf{q}', -\mathbf{q}) \; . \tag{8.12}$$

From Eqs. (8.11) and (8.12), we obtain the following reciprocity relation for the amplitude propagator:

$$\begin{aligned} k_z(q) \int h(\mathbf{r}, \mathbf{r}') \, \exp(-i\mathbf{q} \cdot \boldsymbol{\rho} + i\mathbf{q}' \cdot \boldsymbol{\rho}') \, d^2\rho \, d^2\rho' = \\ k_z(q') \int h(\mathbf{r}', \mathbf{r}) \, \exp(-i\mathbf{q} \cdot \boldsymbol{\rho} + i\mathbf{q}' \cdot \boldsymbol{\rho}') \, d^2\rho \, d^2\rho'. \end{aligned} \tag{8.13}$$

8.4 Coherent backscattering enhancement

In the computation of the reflected diffuse intensity, the reciprocity relation (8.13) induces contributions that are not accounted for in the ladder approximation, and that cannot be neglected even when $k_0\ell_s \gg 1$. Using Eq. (8.13) to transform $h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'})$ into $h^*(\mathbf{r}_{a'}, \mathbf{r}_{b'})$ in Eq. (8.5), we obtain

$$\begin{aligned} I_c(\mathbf{k}_a, \mathbf{k}_b) &= k_z(q_b) k_z(q_a) \, |E_0|^2 \int \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{a'}, \mathbf{r}_{b'}) \rangle \, \exp[i\mathbf{q}_a \cdot (\boldsymbol{\rho}_a - \boldsymbol{\rho}_{a'})] \\ &\quad \times \exp[-i\mathbf{q}_b \cdot (\boldsymbol{\rho}_b - \boldsymbol{\rho}_{b'})] \, d^2\rho_a d^2\rho_b d^2\rho_{a'} d^2\rho_{b'} \; . \end{aligned} \tag{8.14}$$

This expression can be understood as describing the interference between the field produced by a scattering sequence, and the field produced by the reciprocal sequence (same sequence followed in reverse order). This contribution, sometimes refereed to as cooperon, is graphically illustrated in Fig. 8.3. A simple change of variables in Eq. (8.14) allows us to rewrite it in the form

$$\begin{aligned} I_c(\mathbf{k}_a, \mathbf{k}_b) &= k_z(q_b) k_z(q_a) \, |E_0|^2 \int \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'}) \rangle \, \exp[i\mathbf{q}_a \cdot (\boldsymbol{\rho}_a - \boldsymbol{\rho}_{b'})] \\ &\quad \times \exp[-i\mathbf{q}_b \cdot (\boldsymbol{\rho}_b - \boldsymbol{\rho}_{a'})] \, d^2\rho_a d^2\rho_b d^2\rho_{a'} d^2\rho_{b'}. \end{aligned} \tag{8.15}$$

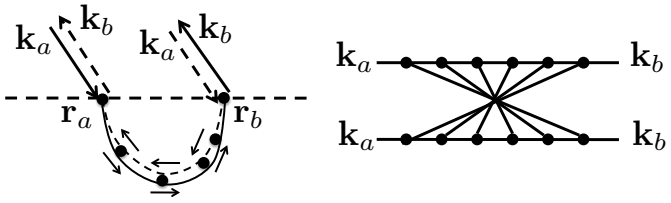

Figure 8.3: Graphical (left) and diagrammatic (right) representations of the reciprocal scattering sequences contributing to coherent backscattering. The diagram on the right is the maximally-crossed diagram.

Using again Eq. (8.6) to simplify the correlator of the amplitude propagator, we find that

$$I_c(\mathbf{k}_a, \mathbf{k}_b) = k_z(q_b)k_z(q_a)\,|E_0|^2 \int P(\boldsymbol{\rho}_b - \boldsymbol{\rho}_a)\,\exp[i(\mathbf{q}_a + \mathbf{q}_b)\cdot(\boldsymbol{\rho}_a - \boldsymbol{\rho}_b)]d^2\rho_a d^2\rho_b. \quad (8.16)$$

This expression describes a contribution to the diffuse reflected intensity that cannot be neglected in the vicinity of the backscattering direction. Indeed, in the exact backscattering direction defined by $\mathbf{k}_b = -\mathbf{k}_a$, Eq. (8.16) is identical to Eq. (8.7), so that:

$$I_c(\mathbf{k}_a, \mathbf{k}_b = -\mathbf{k}_a) = I_d(\mathbf{k}_a, \mathbf{k}_b = -\mathbf{k}_a)\ . \quad (8.17)$$

Therefore, the backscattered diffuse intensity is twice the value predicted in the ladder approximation. This is a consequence of wave reciprocity. This phenomenon, known as coherent backscattering, results from a constructive interference between the fields scattered along reciprocal scattering sequences. Reciprocity ensures that in the exact backscattering direction, these two fields have identical phases, thus producing a constructive interference.

8.5 Coherent backscattering cone and angular width

It is possible to evaluate the lineshape of the intensity distribution around the backscattering direction $\mathbf{k}_b = -\mathbf{k}_a$. Let us introduce the change of variables $\mathbf{X} = \boldsymbol{\rho}_a - \boldsymbol{\rho}_b$ and $\boldsymbol{\rho} = (\boldsymbol{\rho}_a + \boldsymbol{\rho}_b)/2$ (with unit Jacobian) into Eq. (8.16). We obtain

$$I_c(\mathbf{k}_a, \mathbf{k}_b) = k_z(q_b)k_z(q_a)\,|E_0|^2 \left[\int d^2\rho\right]\ \tilde{P}(\mathbf{q}_a + \mathbf{q}_b) \quad (8.18)$$

where $\tilde{P}(\mathbf{q})$ is the Fourier transform of the transport probability. The remaining integral is in practice not infinite, and corresponds to the size S of the illuminated region on the interface. We end up with

$$I_c(\mathbf{k}_a, \mathbf{k}_b) = k_z(q_b)k_z(q_a)\,S\,|E_0|^2\,\tilde{P}(\mathbf{q}_a + \mathbf{q}_b) \quad (8.19)$$

showing that the coherent backscattering intensity I_c is proportional to the Fourier transform of the transport probability $P(\mathbf{r}-\mathbf{r}')$ connecting two points $\mathbf{r}$ and $\mathbf{r}'$ on the surface.

The transport probability can be evaluated using the diffusion approximation. For a non-absorbing semi-infinite medium, its expression can be calculated using the formalism of chapter 5. Assuming $q\ell_t \ll 1$ in the diffusive regime, the calculation (not shown here, see for example Refs. [11, 1] for solutions of the diffusion equation in simple geometries) leads to

$$\tilde{P}(\mathbf{q}) \simeq A[1-(2/3)q\ell_t] \tag{8.20}$$

where A is a constant that we do not specify. Inserting this result into Eq. (8.19) leads to

$$I_c(\mathbf{k}_a,\mathbf{k}_b) = k_z(q_b)k_z(q_a)\,S\,|E_0|^2 A[1-(2/3)\delta q\,\ell_t] \tag{8.21}$$

where we have written $\mathbf{q}_b = -\mathbf{q}_a+\delta\mathbf{q}$, and $\delta q = |\delta\mathbf{q}|$ (see Fig. 8.4). This can also be rewritten as

$$I_c(\mathbf{k}_a,\mathbf{k}_b) \simeq I_c(\delta q=0)[1-(2/3)\delta q\,\ell_t] \tag{8.22}$$

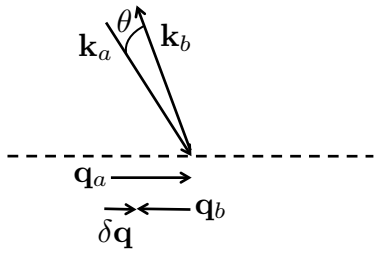

Figure 8.4: Wavevectors involved in the computation of the backscattered diffuse intensity.

Equation (8.21) describes the angular dependence of the reflected intensity around the backscattering direction $\delta q = 0$. Writing $\delta q \simeq k_0\theta$, with θ the angular deviation from the exact backscattering direction, we see that the backscattering intensity peak has an angular width $\Delta\theta \simeq \lambda/\ell_t$. We also see that the backscattering peak exhibits a triangular singularity. This singularity is a signature of the long scattering paths involved in the multiple scattering process. In the presence of absorption, the contribution of these long paths is reduced, and both the amplitude and the sharpness of the backscattering peak decrease. All these features have been observed experimentally. An example of a measured backscattering peak in a non-absorbing medium is shown in Fig. 8.5. The factor of two in the enhancement, the triangular shape of the backscattering cone and the dependence of the angular width on ℓ_t are clearly visible.

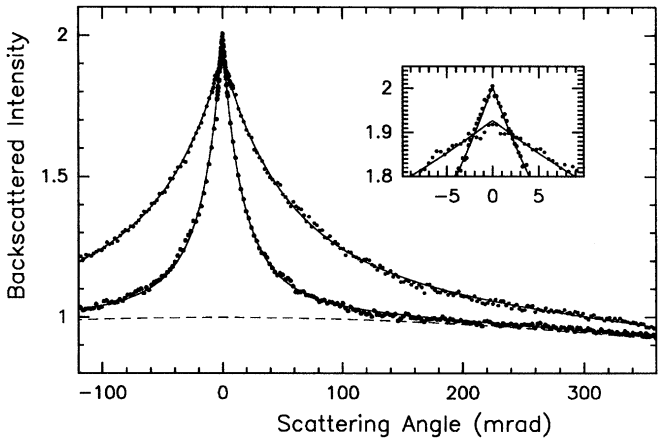

Figure 8.5: Experimental evidence of coherent backscattering of light from a strongly scattering medium. Narrow cone: $k\ell_t \simeq 23$. Wide cone: $k\ell_t \simeq 6$ (with k the wavenumber in the medium). From D. Wiersma *et al.*, Phys. Rev. Lett. **74**, 4193 (1995).

Chapter 9

Angular speckle correlations

In this chapter we study the far-field angular correlation function of the intensity in a speckle pattern produced in transmission through a slab of scattering material. We restrict the discussion to short-range correlations in speckles obeying Gaussian statistics, and focus on practical implications.

9.1 Definition of the angular correlation function

With reference to the geometry in Fig. 9.1, we study the speckle pattern produced in transmission through a slab of thickness L, assumed to be infinite along the transverse directions Ox and Oy.

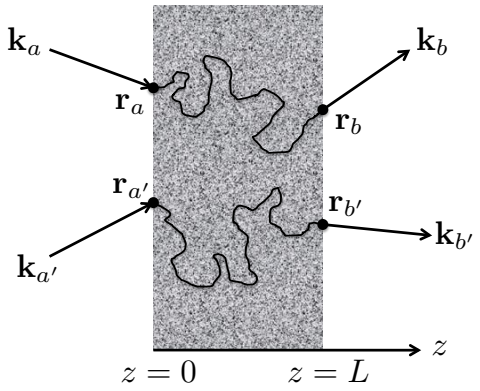

Figure 9.1: Geometry used for the calculation of the angular correlation function of the intensity transmitted through a slab of scattering material.

In the analysis of a far-field speckle pattern, a useful quantity is the correlation function between the intensity $I(\mathbf{k}_a, \mathbf{k}_b)$ emerging in direction $\mathbf{k}_b$ when the medium is illuminated by a plane wave with wavevector $\mathbf{k}_a$, and the intensity $I(\mathbf{k}_{a'}, \mathbf{k}_{b'})$ observed in direction $\mathbf{k}_{b'}$ with an illumination from direction $\mathbf{k}_{a'}$. We use specific notations for the projection of vectors along the (Ox, Oy) plane, such that $\mathbf{r}_a = (\boldsymbol{\rho}_a, z = 0)$, $\mathbf{r}_b = (\boldsymbol{\rho}_b, z = L)$, $\mathbf{k}_a = [\mathbf{q}_a, k_z(q_a)]$, etc.

The angular correlation function of the intensity fluctuations $\delta I = I - \langle I \rangle$ is defined as

$$C^I_{aba'b'} = \frac{\langle \delta I(\mathbf{k}_a, \mathbf{k}_b)\, \delta I(\mathbf{k}_{a'}, \mathbf{k}_{b'}) \rangle}{\langle I(\mathbf{k}_a, \mathbf{k}_b) \rangle \langle I(\mathbf{k}_{a'}, \mathbf{k}_{b'}) \rangle} \ . \tag{9.1}$$

In the regime $k_0 \ell_s \gg 1$, with $k_0 = \omega/c = 2\pi/\lambda$ and ℓ_s the scattering mean free path, we can assume that the field obeys Gaussian statistics (the speckle is fully developped, as discussed in chapter 6). As a result, the intensity correlation function can be factorized as the square of the field correlation function:[1]

$$C^I_{aba'b'} = |C^E_{aba'b'}|^2 \tag{9.2}$$

and we are left with the determination of the normalized angular correlation function of the field, that reads

$$C^E_{aba'b'} = \frac{\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'}) \rangle}{\sqrt{\langle |E(\mathbf{k}_a, \mathbf{k}_b)|^2 \rangle} \sqrt{\langle |E(\mathbf{k}_{a'}, \mathbf{k}_{b'})|^2 \rangle}} \ . \tag{9.3}$$

In this expression we have denoted by $E(\mathbf{k}_a, \mathbf{k}_b)$ the far-field amplitude of the scattered field in direction $\mathbf{k}_b$, for an illumination by a plane wave with wavevector $\mathbf{k}_a$.

9.2 Field angular correlation function in transmission

The scattered field $E(\mathbf{k}_a, \mathbf{k}_b)$ can be written in terms of the amplitude propagator $h(\mathbf{r}_b, \mathbf{r}_a)$ introduced in chapter 3. For an incident plane wave $E_0 \exp(i\mathbf{k}_a \cdot \mathbf{r})$, the field emerging at point $\mathbf{r}_b$ on the exit surface $z = L$ is

$$E(\boldsymbol{\rho}_b, z = L) = \int_{z=0} h(\mathbf{r}_b, \mathbf{r}_a)\, E_0\, \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a)\, d^2\rho_a \tag{9.4}$$

where $\mathbf{r}_a = (\boldsymbol{\rho}_a, z = 0)$ and $\mathbf{r}_b = (\boldsymbol{\rho}_b, z = L)$ are the input and output points. The far-field scattered in direction $\mathbf{k}_b$ takes the form (see for example Ref. [24, 1])

$$E(\mathbf{r}) = \frac{k_z(q_b)}{2i\pi}\, E(\mathbf{q}_b)\, \frac{\exp(ikr)}{r} \tag{9.5}$$

where $k_z(q) = (k_0^2 - \mathbf{q}^2)^{1/2}$ is the z-component of the wavevector $\mathbf{k}$ and $E(\mathbf{q}_b)$ is the Fourier transform of the scattered field in the plane $z = L$:

$$E(\mathbf{q}_b) = \int_{z=L} E(\boldsymbol{\rho}_b, z = L)\, \exp(-i\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_b \ . \tag{9.6}$$

[1] The factorization is the same as that leading to the Siegert relation derived in chapter 7, section 7.3.

Identifying $E(\mathbf{k}_a, \mathbf{k}_b)$ and $k_z(q_b)E(\mathbf{q}_b)$ leads to

$$E(\mathbf{k}_a, \mathbf{k}_b) = k_z(q_b)\, E_0 \int_{z=0} \int_{z=L} h(\mathbf{r}_b, \mathbf{r}_a)\, \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a - i\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_a\, d^2\rho_b\ . \tag{9.7}$$

This is the expression of the scattered far field. In practice the far-field conditions are met in the focal plane of a converging lens, or in the Fourier plane of a microscope objective.

From this expression, the angular correlation function of the field is readily deduced:

$$\begin{aligned}\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle = k_z(q_b)k_z(q_{b'})\, |E_0|^2 \int_{z=0} \int_{z=L} \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'})\rangle \\ \times \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a - i\mathbf{q}_b \cdot \boldsymbol{\rho}_b - i\mathbf{q}_{a'} \cdot \boldsymbol{\rho}_{a'} + i\mathbf{q}_{b'} \cdot \boldsymbol{\rho}_{b'})\, d^2\rho_a d^2\rho_b d^2\rho_{a'} d^2\rho_{b'}\ .\end{aligned} \tag{9.8}$$

The integrals sum up all entry points $\boldsymbol{\rho}_a$ and $\boldsymbol{\rho}'_a$ and exit points $\boldsymbol{\rho}_b$ and $\boldsymbol{\rho}'_b$ (see Fig. 9.1). In the ladder approximation, that has been introduced qualitatively in chapter 3, the correlator of the amplitude propagator can be simplified into (see Eq. 6.7):

$$\langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_{b'}, \mathbf{r}_{a'})\rangle \simeq P(\mathbf{r}_b, \mathbf{r}_a)\, \delta(\mathbf{r}_a - \mathbf{r}_{a'}) \delta(\mathbf{r}_b - \mathbf{r}_{b'}) \tag{9.9}$$

where $P(\mathbf{r}_b, \mathbf{r}_a)$ is the intensity propagator from $\mathbf{r}_a$ to $\mathbf{r}_b$. In the geometry considered here, due to translationnal invariance along the transverse directions, $P(\mathbf{r}_b, \mathbf{r}_a)$ depends on $\mathbf{r}_a$ and $\mathbf{r}_b$ only through the difference $\boldsymbol{\rho}_b - \boldsymbol{\rho}_a$, and we can rewrite Eq. (9.8) in the form

$$\begin{aligned}\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle &= k_z(q_b)k_z(q_{b'})\, |E_0|^2 \int_{z=0} \int_{z=L} P(\boldsymbol{\rho}_b - \boldsymbol{\rho}_a) \\ &\quad \times \exp(i\Delta\mathbf{q}_a \cdot \boldsymbol{\rho}_a - i\Delta\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_a d^2\rho_b.\end{aligned} \tag{9.10}$$

We have introduced the notations $\Delta\mathbf{q}_a = \mathbf{q}_a - \mathbf{q}_{a'}$ and $\Delta\mathbf{q}_b = \mathbf{q}_b - \mathbf{q}_{b'}$ for clarity. In order to simplify the integrals, we perform the change of variables $\mathbf{X} = \boldsymbol{\rho}_a - \boldsymbol{\rho}_b$ and $\boldsymbol{\rho} = (\boldsymbol{\rho}_a + \boldsymbol{\rho}_b)/2$ (with unit Jacobian), leading to

$$\begin{aligned}\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle &= k_z(q_b)k_z(q_{b'})\, |E_0|^2 \int P(\mathbf{X})\, \exp[i(\Delta\mathbf{q}_a + \Delta\mathbf{q}_b) \cdot \mathbf{X}/2]\, d^2X \\ &\quad \times \int \exp[i(\Delta\mathbf{q}_a - \Delta\mathbf{q}_b) \cdot \boldsymbol{\rho}]\, d^2\rho.\end{aligned} \tag{9.11}$$

The first integral is the Fourier transform $\tilde{P}(\mathbf{q})$ of $P(\mathbf{X})$, calculated for $\mathbf{q} = (\Delta\mathbf{q}_a + \Delta\mathbf{q}_b)/2$. The second integral is the Dirac distribution $4\pi^2\, \delta(\Delta\mathbf{q}_a - \Delta\mathbf{q}_b)$. We finally end up with a simple expression for the field correlation function:

$$\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle = k_z(q_b)k_z(q_{b'})\, |E_0|^2\, \tilde{P}(\Delta\mathbf{q}_a)\, \delta(\Delta\mathbf{q}_a - \Delta\mathbf{q}_b). \tag{9.12}$$

This result shows that the angular correlation function of the field is different from zero only when $\Delta\mathbf{q}_a = \Delta\mathbf{q}_b$. Moreover, when $\Delta q_a = |\Delta\mathbf{q}_a|$ increases, the range of the correlation is described by the Fourier transform $\tilde{P}(\Delta\mathbf{q}_a)$ of the intensity propagator.

9.3 Intensity propagator in the diffusion approximation

Considering a slab with thickness $L \gg \ell_t$, the intensity propagator can be calculated using the diffusion approximation. Consider the solution of the diffusion equation in a non absorbing medium and a delta function source term:

$$\nabla^2_{\mathbf{r}_a} L(\mathbf{r}_b, \mathbf{r}_a) = -\delta(\mathbf{r}_b - \mathbf{r}_a) \tag{9.13}$$

with appropriate boundary conditions on the slab surfaces. In other words, $L(\mathbf{r}_b, \mathbf{r}_a)$ is the Green function of the diffusion equation. This equation in a slab geometry can be solved in Fourier space, using the boundary conditions described in Appendix **??**. The calculation is not detailed here (see for example Ref. [11] for solutions of the diffusion equation in simple geometries). Writing $\tilde{L}(\mathbf{q}, z, z')$ the Fourier transform of the propagator $L(\boldsymbol{\rho}_b - \boldsymbol{\rho}_a, z, z')$ with respect to $\boldsymbol{\rho}_b - \boldsymbol{\rho}_a$, we obtain

$$\tilde{L}(\mathbf{q}, z = L, z' = 0) = \frac{q z_0^2}{\sinh(qL) + 2qz_0\,\cosh(qL) + (qz_0)^2 \sinh(qL)}\ , \tag{9.14}$$

where $q = |\mathbf{q}|$ and $z_0 = (2/3)\ell_t$ is the extrapolation distance that appears in the boundary conditions (see Appendix **??**). In the regime $q\ell_t \ll 1$, the intensity propagator, that is proportionnal to $\tilde{L}$, takes the form

$$\tilde{P}(\mathbf{q}) = A\,\frac{q}{\sinh(qL)} \tag{9.15}$$

where A is a prefactor that we do not need to specify.

9.4 Intensity correlation function and memory effect

An explicit expression of the field correlation function is obtained by inserting (9.15) into (9.12), leading to

$$\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle = A\, k_z(q_b) k_z(q_{b'})\, |E_0|^2\, \frac{\Delta q_a}{\sinh(\Delta q_a\, L)}\, \delta(\Delta\mathbf{q}_a - \Delta\mathbf{q}_b). \tag{9.16}$$

The normalized correlation function of the intensity can be deduced using Eqs. (9.2) and (9.3). It can be formally written as

$$C^I_{aba'b'} = \left|\frac{\Delta q_a\, L}{\sinh(\Delta q_a\, L)}\right|^2 \delta_{\Delta\mathbf{q}_a, \Delta\mathbf{q}_b} \tag{9.17}$$

where $\delta_{\Delta\mathbf{q}_a, \Delta\mathbf{q}_b}$ is a Kronecker delta.

This angular correlation function describes the so-called "memory effect". Indeed, it shows that by changing the angle of incidence from $\mathbf{q}_a$ to $\mathbf{q}_{a'} = \mathbf{q}_a + \Delta\mathbf{q}_a$, the speckle pattern observed in direction $\mathbf{q}_{b'} = \mathbf{q}_b + \Delta\mathbf{q}_a$ remains correlated to the initial speckle pattern observed in direction $\mathbf{q}_b$ (the speckle pattern seems to move as a whole). This effect remains visible as long as the amplitude of the correlation does not vanish when Δq_a increases. When the condition $\Delta q_a L \gg 1$ is satisfied, $C^I_{aba'b'} \sim \exp(-2\Delta q_a L)$, showing that the angular intensity correlation function calculated in the ladder approximation is a short-range correlation function.

9.5 Size of a speckle spot

In a speckle pattern, one observes a complex distribution of bright and dark spots (see Fig. 6.1 in chapter 6). The intensity correlation function can be used to characterize the typical size of a speckle spot.

To address this question, we still consider the transmission geometry in Fig. 9.1, but with an illuminating beam of finite transverse size. In the paraxial approximation, the complex amplitude of the field produced by such a beam in the plane $z = 0$ can be written formally as $E_0(\boldsymbol{\rho}_a)\, \exp(i\mathbf{q}_a \cdot \boldsymbol{\rho}_a)$. In these conditions, Eq. (9.10) is transformed into

$$\begin{aligned} \langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle &= k_z(q_b) k_z(q_{b'}) \int_{z=0} \int_{z=L} H(\boldsymbol{\rho}_a)\, P(\boldsymbol{\rho}_b - \boldsymbol{\rho}_a) \\ &\times \exp(i\Delta\mathbf{q}_a \cdot \boldsymbol{\rho}_a - i\Delta\mathbf{q}_b \cdot \boldsymbol{\rho}_b)\, d^2\rho_a d^2\rho_b \end{aligned} \tag{9.18}$$

where $H(\boldsymbol{\rho}_a) = |E_0(\boldsymbol{\rho}_a)|^2$ is the intensity distribution of the beam in the input plane $z = 0$. Using again the same change of variables as that leading to Eq. (9.11), and performing the Fourier transforms, leads to

$$\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle = k_z(q_b) k_z(q_{b'})\, \tilde{P}(\Delta\mathbf{q}_b)\, \tilde{H}(\Delta\mathbf{q}_b - \Delta\mathbf{q}_a)\ . \tag{9.19}$$

A measure of the angular size of a speckle spot is the width of the correlation function (9.19), considered as a function of the observation direction $\mathbf{q}_b$, and for a fixed direction of incidence $\mathbf{q}_a$ (in practice one usually measures the width of the intensity correlation function, that is essentially the square modulus of the field correlation function). We therefore need to evaluate the width of $\langle E(\mathbf{k}_a, \mathbf{k}_b)\, E^*(\mathbf{k}_{a'}, \mathbf{k}_{b'})\rangle$ considered as a function of $\Delta\mathbf{q}_b$ with $\Delta\mathbf{q}_a = 0$. The result depends on the respective widths of the two functions in the right-hand side in Eq. (9.19).

For a slab with thickness $L \gg \ell_t$, the solution of the diffusion equation shows that $P(\boldsymbol{\rho}_a - \boldsymbol{\rho}_b)$ leads to a spatial distribution of diffuse intensity of size L in the outpout plane $z = L$. The function $H(\boldsymbol{\rho}_a)$ has a width W, corresponding to the beam size in the plane $z = 0$. Two different situations have to be considered.

Extended beam $(W \gg L)$

In this case the angular width of the correlation function is driven by the function $\tilde{H}(\Delta\mathbf{q}_b)$. This width is given by $\Delta q_b \sim 2\pi/W$. If one observes the speckle pattern in the focal plane of an imaging system with image focal length f, the size of the speckle spot is $\Delta R \sim f\lambda/W$.

Focussed illumination $(W \ll L)$

In this case the angular width of the correlation function is driven by the function $\tilde{P}(\Delta\mathbf{q}_b)$. As discussed in the previous section, this width is $\Delta q_b \sim 2\pi/L$. This gives a speckle spot size $\Delta R \sim f\lambda/L$ in the focal plane of an imaging system.

9.6 Number of transmission modes

In the case of an illumination with a beam of finite transverse size, the angular size of a speckle spot can be associated with the size of a transmitted mode. This assertion is based on the intuitive picture that two transmitted wavevectors will be independent (and will describe two different modes) when their angular separation is larger than the angular range of the intensity correlation function.

Let us denote by θ the angle between two transmitted wavevectors $\mathbf{k}_b$ and $\mathbf{k}_{b'}$. We have $|\Delta\mathbf{q}_b|^2 \simeq |\Delta\mathbf{k}_b|^2 = 2k_0^2\,(1-\cos\theta)$. For a beam of width W satisfying $W \gg L$, we have $|\Delta\mathbf{q}_b| \simeq 2\pi/W$. Therefore the angle θ defines the angular extent of a mode when

$$2k_0^2\,(1-\cos\theta) \simeq \frac{4\pi^2}{W^2} \tag{9.20}$$

The angle θ also corresponds to a solid angle through the relation $\Delta\Omega = 2\pi(1-\cos\theta)$, so that a transmission mode corresponds to a solid angle $\Delta\Omega \simeq \pi\lambda^2/W^2$. The number of transmission modes is evaluated as

$$N_{\rm modes} = \frac{2\pi}{\Delta\Omega} \simeq \frac{2\,W^2}{\lambda^2} \tag{9.21}$$

This number of modes is analogous to the number of transverse modes that is used in transport of waves through waveguides, as in mesoscopic electronic transport. It is also a reliable evaluation of the number of degrees of freedom that are available to act on light transmission through a disordered medium in the multiple scattering regime.

Part IV

Complements

Complement A

Scattering matrix for polarized light

In this complement we briefly review the description of the scattering matrix for polarized light in terms of the Stokes vector.

Scattering matrix

We take the incident plane wave to be of the form

$$\mathbf{E}_0(\mathbf{r}) = \mathbf{E}_0 \, \exp(ik_0 \mathbf{u}_{inc} \cdot \mathbf{r}) \, ,$$

where $\mathbf{E}_0 = E_0 \, \mathbf{e}_0$, with the unit vector $\mathbf{e}_0$ describing the direction of polarization, and $\mathbf{u}_{inc}$ is the unit vector defining the propagation direction. In terms of the scattering matrix introduced in chapter 2, the far-field amplitude of the scattered field is

$$\mathbf{E}_s(\mathbf{r}) = \mathbf{S}(\mathbf{u}) \, E_0 \, \mathbf{e}_0 \frac{\exp(ik_0 r)}{r} \, . \tag{A.1}$$

Technically, $\mathbf{S}(\mathbf{u})$ is a second-rank tensor (that transforms a vector into another vector). Indeed, there is no reason for $\mathbf{E}_s$ and $\mathbf{E}_0$ to be colinear.

The polarization state of the incident and scattered fields can be defined using two vector components (the fields are transverse). To proceed, we need to define a reference plane, and decompose the fields $\mathbf{E}_0$ and $\mathbf{E}_s$ into their parallel($\|$) and perpendicular ($\perp$) components. The reference plane is defined using the incidence direction (chosen to coincide with the z-axis) and the scattering direction, as shown in Fig. A.1:

$$\mathbf{E}_0 = E_0^{\|} \, \mathbf{e}_i^{\|} + E_0^{\perp} \, \mathbf{e}_i^{\perp}$$

$$\mathbf{E}_s = E_s^{\|} \, \mathbf{e}_s^{\|} + E_s^{\perp} \, \mathbf{e}_s^{\perp} \, .$$

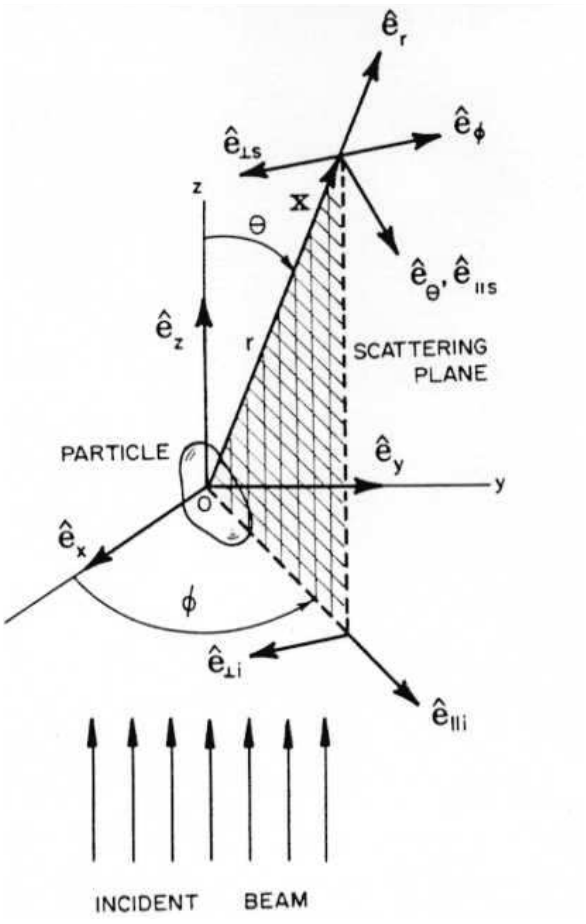

Figure A.1: Geometry used to define the scattering matrix for polarized light. From Ref. [2].

Using these bases, the scattering matrix $\mathbf{S}(\mathbf{u})$ is usually written in the form [2]

$$\begin{pmatrix} E_s^{\parallel} \\ E_s^{\perp} \end{pmatrix} = \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \frac{\exp(ik_0 r)}{r} \begin{pmatrix} E_0^{\parallel} \\ E_0^{\perp} \end{pmatrix} \tag{A.2}$$

where each element of the scattering matrix is a function of the scattering direction (θ, ϕ), of frequency, and depends on the type of particle.

In the particular case of spherical homogeneous particles, the scattering matrix has the following properties:

- $S_3 = S_4 = 0$
- For forward scattering ($\theta = 0$), $S_1(0) = S_2(0) = S(0)$.

Finally, for electromagnetic waves, the scattering matrix also leads to a simple expression of the optical theorem:

$$\sigma_e = \frac{4\pi}{k_0} \operatorname{Im}[\mathbf{e}_0 \cdot \mathbf{S}(\mathbf{u}_{inc})\, \mathbf{e}_0] \,. \tag{A.3}$$

Except for the projection of the scattering matrix on the direction of polarization of the incident wave, the expression is similar to that obtained for scalar waves. We give the derivation of Eq. (A.3) in chapter 2.

Stokes vector

The scattering matrix contains all information on the scattering process. In optics, we often measure intensities rather than field amplitudes. It is convenient to introduce a description of polarization in terms of intensity measurements. Using the basis defined in Fig. A.1, we have seen that the fields are written in the form $\mathbf{E} = E_\parallel\, \mathbf{e}_\parallel + E_\perp\, \mathbf{e}_\perp$ (this decomposition holds for the incident and the scattered fields). We can define four parameters $[I, Q, U, V]$, constituting the Stokes vector:

$$\begin{aligned} I &= E_\parallel E_\parallel^* + E_\perp E_\perp^* \\ Q &= E_\parallel E_\parallel^* - E_\perp E_\perp^* \\ U &= E_\parallel E_\perp^* + E_\parallel^* E_\perp \\ V &= i(E_\parallel E_\perp^* - E_\parallel^* E_\perp) \,. \end{aligned} \tag{A.4}$$

If we introduce the amplitudes and phases of the parallel and perpendicular components $E_\parallel = a_\parallel \exp(i\delta_\parallel)$ et $E_\perp = a_\perp \exp(i\delta_\perp)$, the Stokes vector of the field $\mathbf{E}$ can be rewritten in the form

$$\begin{aligned} I &= a_\parallel^2 + a_\perp^2 \\ Q &= a_\parallel^2 - a_\perp^2 \\ U &= 2\, a_\parallel\, a_\perp\, \cos(\delta_\parallel - \delta_\perp) \\ V &= 2\, a_\parallel\, a_\perp\, \sin(\delta_\perp - \delta_\parallel) \,. \end{aligned} \tag{A.5}$$

These expressions clearly show that the first two parameters measure the sum and difference of intensity in the two components, while the other parameters measure the relative phases. The Stokes vector contains the information on the relative amplitude and phases of the two vector components of the field (and therefore on the polarization state), although it results only from intensity measurements. More precisely, the four elements can be measured as follows:

- I : Total intensity.

- Q : Difference between the intensity of the $\parallel$ component and the intensity of the $\perp$ component. Can be obtained from two intensity measurements, using a polarizer oriented either along $\mathbf{e}_\parallel$ or along $\mathbf{e}_\perp$.
- U : Difference between two intensities I^+ et I^-. I^+ is measured after a polarizer oriented along the direction $\mathbf{e}_\parallel + \mathbf{e}_\perp$. I^- is measured after a polarizer oriented along the direction $\mathbf{e}_\parallel - \mathbf{e}_\perp$.
- V : Difference between the intensities of the right and left circular polarizations. The connection between linear and circular polarizations is made by using the relation $\mathbf{E} = E_\parallel \, \mathbf{e}_\parallel + E_\perp \, \mathbf{e}_\perp = E_s \, \mathbf{e}_D + E_G \, \mathbf{e}_G$ where the vectors defining the right and left circular polarizations are $\mathbf{e}_D = (\mathbf{e}_\parallel + i\mathbf{e}_\perp)/\sqrt{2}$ and $\mathbf{e}_G = (\mathbf{e}_\parallel - i\mathbf{e}_\perp)/\sqrt{2}$.

Mueller matrix

In a scattering configuration, we can define a Stokes vector for both the incident and the scattered fields. Using the definition of the scattering matrix, we can show that a linear relation exists between the two Stokes vectors. The matrix that describes this linear relation is known as the Mueller matrix. It is usually written in the following form [2]:

$$\begin{bmatrix} I_d \\ Q_d \\ U_d \\ V_d \end{bmatrix} = \begin{bmatrix} S_{11} & .. & S_{14} \\ . & & . \\ . & & . \\ S_{41} & .. & S_{44} \end{bmatrix} \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix} \tag{A.6}$$

The concepts of Stokes vectors and Mueller matrix are useful to describe the transport of light in complex media, accounting for the polarization degrees of freedom [14].

Complement B

Average field and Dyson equation

In this complement, we summarize the treatment of the average field in multiple scattering theory. In the independent scattering approximation (ISA), we retrieve the decay of the average field in terms of the extinction mean free path, as derived in chapter 3 using another approach.

Dyson equation

Starting from Eq. (3.6), we immediately find that the average field satisfies

$$\langle E \rangle = E_0 + G_0 \langle T \rangle E_0 \,, \tag{B.1}$$

since T is the only random object in the right-hand side (the incident field E_0 and the free-space Green's function are deterministic). Inverting this equation, we find that $E_0 = (I + G_0\langle T\rangle)^{-1}\langle E\rangle$, which, after insertion in the last term in Eq. (B.1), leads to

$$\langle E \rangle = E_0 + G_0 \, \Sigma \, \langle E \rangle \,. \tag{B.2}$$

In this expression Σ is a new operator, known as the self-energy[1], and given by

$$\Sigma = \langle T \rangle (I + G_0 \langle T \rangle)^{-1} \,. \tag{B.3}$$

The equation for the averaged field can also be written in terms of Green's function. Denoting by $s(\mathbf{r})$ the source of the incident field (as in chapter 1, section 1.1.1), we have

$$E_0(\mathbf{r}) = \int G_0(\mathbf{r}, \mathbf{r}') \, s(\mathbf{r}') \, d^3 r' \,, \tag{B.4}$$

and

$$E(\mathbf{r}) = \int G(\mathbf{r}, \mathbf{r}') \, s(\mathbf{r}') \, d^3 r' \,. \tag{B.5}$$

[1] The name of this operator comes from quantum field theory, for which this formalism was developed.

The second relation above defines the Green's function G in the presence of the scattering medium (while G_0 is the bare Green's function of free space). In terms of Green's functions, Eq. (B.2) becomes

$$\langle G \rangle = G_0 + G_0 \, \Sigma \, \langle G \rangle \, , \tag{B.6}$$

which is knwon as the Dyson equation. Solving this equation for $\langle G \rangle$ is far from being easy since Σ is a complicated object. Nevertheless, the Dyson equation is a fundamental brick in multiple scattering theory.

Weak scattering and independent scattering approximation

In a weakly scattering medium, we can find the expression of the self-energy Σ and examine the behavior of the average field using a perturbative expansion. To lowest order, Eq. (B.3) leads to $\Sigma = \langle T \rangle$. For a medium made of discrete scatterers with T-matrix t_j, as studied in chapter 3, we also find from Eq. (3.14) that to lowest order $T = \sum_j t_j$, which corresponds to the independent scattering approximation. In these conditions, the self-energy is

$$\Sigma = \sum_j \langle t_j \rangle \, . \tag{B.7}$$

Let us consider an ensemble of N subwavelength scatterers distributed in a volume $\mathcal{V}$, and described by their polarizability $\alpha(\omega)$. For a scatterer located at the point $\mathbf{r}_j$, the T-matrix is (see section 3.1.3) $t_j(\mathbf{r}_1, \mathbf{r}_2) = k_0^2 \alpha(\omega) \delta(\mathbf{r}_1 - \mathbf{r}_j) \delta(\mathbf{r}_2 - \mathbf{r}_j)$. The average value is defined as

$$\langle t_j(\mathbf{r}_1, \mathbf{r}_2) \rangle = \int t_j(\mathbf{r}_1, \mathbf{r}_2) \, P(\mathbf{r}_j) \, d^3 r_j \, , \tag{B.8}$$

where $P(\mathbf{r}_j)$ is the probability density of finding a scatterer at the point $\mathbf{r}_j$. For a uniform distribution of scatterers, $P(\mathbf{r}_j) = 1/\mathcal{V}$ and we find that

$$\langle t_j(\mathbf{r}_1, \mathbf{r}_2) \rangle = \frac{1}{\mathcal{V}} k_0^2 \alpha(\omega) \delta(\mathbf{r}_1 - \mathbf{r}_2) \, . \tag{B.9}$$

Using this result in Eq. (B.7), we obtain

$$\Sigma = \rho k_0^2 \alpha(\omega) \delta(\mathbf{r}_1 - \mathbf{r}_2) \, , \tag{B.10}$$

where $\rho = N/\mathcal{V}$ is the average density of scatterers. This is the expression of the self-energy in a weakly scattering ensemble of identical small scatterers.

We can now use this expression to examine the behavior of the average field. In real space Eq. (B.2) reads

$$\langle E(\mathbf{r}) \rangle = E_0(\mathbf{r}) + \int G_0(\mathbf{r}, \mathbf{r}_1) \, \Sigma(\mathbf{r}_1, \mathbf{r}_2) \, \langle E(\mathbf{r}_2) \rangle \, d^3 r_1 d^3 r_2 \, . \tag{B.11}$$

Inserting Eq. (B.10), it becomes

$$\langle E(\mathbf{r})\rangle = E_0(\mathbf{r}) + k_0^2 \int G_0(\mathbf{r},\mathbf{r}')\,\rho\alpha(\omega)\,\langle E(\mathbf{r}')\rangle\,d^3r'\,. \tag{B.12}$$

This equation takes the same form as the Lippmann-Schwinger equation (3.1) with a uniform effective potential $V_{\text{eff}} = k_0^2\rho\alpha(\omega)$, or equivalently in a uniform medium with dielectric function $\epsilon_{\text{eff}} = 1+\rho\alpha(\omega)$. We can note that the weak scattering condition, which amounts to assuming that the effective dielectric function slightly differs from unity, can be written $|\rho\alpha(\omega)| \ll 1$.

Assuming an illumination with a plane wave of the form $E_0(\mathbf{r}) = E_0\exp(ik_0z)$, then the solution $\langle E(\mathbf{r})\rangle$ is a plane wave with wavevector $k_{\text{eff}} = n_{\text{eff}}k_0$, with $n_{\text{eff}} = \sqrt{\epsilon_{\text{eff}}}$. A feature is that the imaginary part of the effective refractive index $\mathrm{Im}n_{\text{eff}}$ induces an exponential decay of the average field. Defining the extinction length ℓ_e as the decay length of the intensity of the average field $|\langle E(\mathbf{r})\rangle|^2 \sim \exp(-z/\ell_e)$, we find that

$$\ell_e = \frac{1}{2k_0\,\mathrm{Im}n_{\text{eff}}}\,. \tag{B.13}$$

Making use of the fact that $\mathrm{Im}n_{\text{eff}} = \mathrm{Im}\sqrt{\epsilon_{\text{eff}}} \simeq \rho\,\mathrm{Im}\alpha(\omega)/2$, and that $k_0\,\mathrm{Im}\alpha(\omega) = \sigma_e$ [see chapter 2 Eq. (2.41)], we obtain

$$\ell_e = \frac{1}{\rho\sigma_e}\,, \tag{B.14}$$

which is the result derived in chapter 3, section 3.3, using a phenomenological approach.

Complement C

Average intensity and Bethe-Salpeter equation

In this complement, we introduce a closed-form equation for the field correlation function known as the Bethe-Salpeter equation. We discuss the ladder approximation that was introduced qualitatively in chapter 3, section 3.5.

Bethe-Salpeter equation

To deal with second-order observables, the most general object is the field correlation function $\langle E(\mathbf{r})E^*(\mathbf{r}')\rangle$. A closed-form equation for this correlation function can be derived, and reads

$$\begin{aligned}\langle E(\mathbf{r})E^*(\mathbf{r}')\rangle &= \langle E(\mathbf{r})\rangle\langle E^*(\mathbf{r}')\rangle \\ &+ \int \langle G(\mathbf{r},\mathbf{r}_1)\rangle\langle G^*(\mathbf{r}',\mathbf{r}_2)\rangle\, \Gamma(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2,\mathbf{r}_4)\, \langle E(\mathbf{r}_3)E^*(\mathbf{r}_4)\rangle\, d^3r_1 d^3r_2 d^3r_3 d^3r_4\,.\end{aligned} \tag{C.1}$$

In this expression Γ is the so-called irreducible vertex, that plays a similar role for the correlation function as the self-energy for the average field (see Complement B). The derivation of this equation is beyond the scope of this lecture. The interested reader can find derivations in textbooks dedicated to multiple scattering of waves [1, 9, 10, 11]. The average Green's function appearing in the integral is the solution to the Dyson equation introduced in Complement B. In operator form the Bethe-Salpeter equation is

$$\langle EE^*\rangle = \langle E\rangle\langle E^*\rangle + \langle G\rangle\langle G^*\rangle\,\Gamma\,\langle EE^*\rangle\,. \tag{C.2}$$

Weak scattering and ladder approximation

Consider a medium made of N discrete scatterers with T-matrix t_j distributed in a volume $\mathcal{V}$, as studied in chapter 3. Assuming weak and independent scattering, we can find the simplest form of the irreducible vertex by keeping only the lowest order in a pertubative expansion. We find that (see for example Ref. [1], chap. 18)

$$\Gamma = \sum_{j=1}^{N} \langle t_j t_j^* \rangle \,. \tag{C.3}$$

This lowest order approximation corresponds to the ladder approximation discussed qualitatively in chapter 3, section 3.5. To make it more explicit, let us calculate Γ for subwavelength scatterers described by their polarizability $\alpha(\omega)$. We have

$$\Gamma(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2,\mathbf{r}_4) = \sum_{j=1}^{N} \langle t_j(\mathbf{r}_1,\mathbf{r}_3) t_j^*(\mathbf{r}_2,\mathbf{r}_4)\rangle = \sum_{j=1}^{N} \int t_j(\mathbf{r}_1,\mathbf{r}_3) t_j^*(\mathbf{r}_2,\mathbf{r}_4)\, P(\mathbf{r}_j)\, d^3 r_j \,, \tag{C.4}$$

where $P(\mathbf{r}_j)$ is the probability density of finding a scatterer at the point $\mathbf{r}_j$. For a uniform distribution of scatterers, $P(\mathbf{r}_j) = 1/\mathcal{V}$ and we find that

$$\Gamma(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2,\mathbf{r}_4) = \rho \int t_j(\mathbf{r}_1,\mathbf{r}_3) t_j^*(\mathbf{r}_2,\mathbf{r}_4)\, d^3 r_j \,, \tag{C.5}$$

where $\rho = N/\mathcal{V}$ is the average density of scatterers. The T-matrix of a scatterer located at the point $\mathbf{r}_j$ is $t_j(\mathbf{r},\mathbf{r}') = k_0^2 \alpha(\omega) \delta(\mathbf{r}-\mathbf{r}_j)\delta(\mathbf{r}'-\mathbf{r}_j)$, which leads to

$$\Gamma(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2,\mathbf{r}_4) = \rho k_0^4 |\alpha(\omega)|^2\, \delta(\mathbf{r}_1-\mathbf{r}_3)\delta(\mathbf{r}_2-\mathbf{r}_4)\delta(\mathbf{r}_1-\mathbf{r}_2) \,. \tag{C.6}$$

For subwavelength scatterers, we have seen in chapter 2 that $k_0^4/(6\pi)|\alpha(\omega)|^2$ is the scattering cross section of a subwavelength scatterer for vector waves (Eq. 2.39). For scalar waves, as considered here, we would find $\sigma_s = k_0^4/(4\pi)|\alpha(\omega)|^2$. Since, the scattering mean free path for independent scattering is such that $1/\ell_s = \rho\sigma_s$, the expression above can also be written

$$\Gamma(\mathbf{r}_1,\mathbf{r}_3;\mathbf{r}_2,\mathbf{r}_4) = \frac{4\pi}{\ell_s} \delta(\mathbf{r}_1-\mathbf{r}_3)\delta(\mathbf{r}_2-\mathbf{r}_4)\delta(\mathbf{r}_1-\mathbf{r}_2) \,. \tag{C.7}$$

Average intensity

Rewriting Eq. (C.2) for $\mathbf{r} = \mathbf{r}'$, and making use of Eq. (C.7), we find an integral equation for the average intensity:

$$\langle I(\mathbf{r})\rangle = |\langle E(\mathbf{r})\rangle|^2 + \frac{4\pi}{\ell_s} \int |\langle G(\mathbf{r},\mathbf{r}_1)\rangle|^2 \, \langle I(\mathbf{r}_1)\rangle \, d^3 r_1 \,. \tag{C.8}$$

From the splitting introduced in chapter 3, section 3.2, we recognize that the first term in the right-hand side is the ballistic intensity I_b, while the intergral describes the diffuse intensity I_d.

An explicit expression of the average intensity can be obtained by iterating Eq. (C.8):

$$\begin{aligned}\langle I(\mathbf{r})\rangle &= I_b(\mathbf{r}) + \int |\langle G(\mathbf{r},\mathbf{r}_1)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) I_b(\mathbf{r}_1)\, d^3r_1 \\ &+ \int |\langle G(\mathbf{r},\mathbf{r}_2)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) |\langle G(\mathbf{r}_2,\mathbf{r}_1)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) I_b(\mathbf{r}_1)\, d^3r_1 d^3r_2 \\ &+ \int |\langle G(\mathbf{r},\mathbf{r}_3)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) |\langle G(\mathbf{r}_3,\mathbf{r}_2)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) |\langle G(\mathbf{r}_2,\mathbf{r}_1)\rangle|^2 \left(\frac{4\pi}{\ell_s}\right) I_b(\mathbf{r}_1)\, d^3r_1 d^3r_2 d^3r_3 \\ &+ \ldots \end{aligned} \tag{C.9}$$

This expression gives a clear picture to the building of the average intensity in the ladder approximation: A series of scattering events with ballisitic propagation of the intensity between them, as illustrated in Fig. C.1. This gives a more rigorous basis to the qualitative description of intensity propagation introduced in chapter 3, section 3.5.

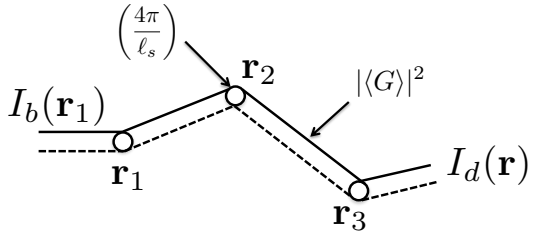

Figure C.1: Illustrating the building of the diffuse intensity in the ladder approximation.

Complement D

Diffuse transmission through a slab

In this complement, we address the question of the boundary conditions at a flat interface between a scattering medium and a non scattering medium (as vacuum or air in optics) when the interface is illuminated by a plane wave at normal incidence (see Fig. D.1). Close to the boundary, the conversion of the incident intensity into diffuse intensity occurs in a layer with a thickness on the order of $\ell_s = 1/\mu_s$. This generalizes the approach introduced in chapter 5 section 5.6. As an application, we calculate the transmission factor for the diffuse intensity propagating through a scattering slab.

Diffusion equation with collimated illumination

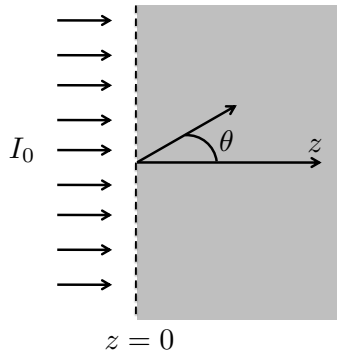

Figure D.1: Geometry used to study the boundary conditions in the diffusion approximation. The medium $z < 0$ is non scattering, while the medium $z > 0$ is assumed to be strongly scattering. The interface $z = 0$ is illuminated by a plane wave with intensity I_0 (W.m^{-2}).

In chapter 4, we have seen that the diffuse specific intensity obeys a RTE with a source

term resulting from the ballisitic intensity (Eq. 4.14). In steady-state, and in the geometry of Fig. D.1 in which the specific intensity only depends on z and $\mathbf{u}$ due to translationnal invariance along the $x-y$ directions, we have

$$\begin{aligned}\mathbf{u}\cdot\frac{\partial}{\partial z}I_{\rm d}(z,\mathbf{u})\,\mathbf{e}_z &= -(\mu_a+\mu_s)\,I_{\rm d}(z,\mathbf{u})+\frac{\mu_s}{4\pi}\int_{4\pi}p(\mathbf{u}\cdot\mathbf{u}')\,I_{\rm d}(z,\mathbf{u}')\,d\Omega' \\ &\quad +\frac{\mu_s}{4\pi}\,p(\mathbf{u}\cdot\mathbf{e}_z)\,I_{\rm b}(z)\;. \end{aligned} \tag{D.1}$$

Here the ballistic intensity is $I_{\rm b}(z)=I_0\,\exp[-(\mu_a+\mu_s)z]$, where I_0 is the intensity of the incident wave (for simplicity we assume that both media are index matched so that here is no specular reflection on the interface). We have used the notation $\mathbf{e}_z$ for the unit vector along the z direction.

Following the same procedure as in chapter 5, we can start from Eq. (D.1) and derive a diffusion equation valid at large scales. After some technical calculations (that are left as an exercise), we obtain a diffusion equation for the energy density $U(z)$, with a source term resulting from the ballistic incident intensity:

$$\frac{d^2\,U}{dz^2}-3\mu_a(\mu_a+\mu_s')\,U(z)=-\frac{3\mu_s(\mu_a+\mu_s')}{v_E}\,I_{\rm b}(z)-\frac{3\mu_s g(\mu_a+\mu_s)}{v_E}\,I_{\rm b}(z)\;. \tag{D.2}$$

Here we use the notation $\mu_s'=\mu_s(1-g)=1/\ell_t$ (sometimes denoted by reduced scattering coefficient). Remember that since we have used the P_1 approximation (see chapter 5), this expression is *a priori* valid in the regime $\mu_a\ll\mu_s'$. The energy current is $\mathbf{q}(z)=q(z)\,\mathbf{e}_z$ with

$$q(z)=-\frac{v_E}{3(\mu_a+\mu_s')}\,\frac{dU}{dz}+\frac{\mu_s\,g}{\mu_a+\mu_s'}\,I_{\rm b}(z)\;. \tag{D.3}$$

Boundary condition at $z=0$

In order to get a boundary condition in terms of the energy density, we use the fact that the diffuse incoming flux vanishes at the interface, as in chapter 5, following the approach introduced in Ref. [16]. In terms of the diffuse specific intensity, this reads as

$$\int_{2\pi}I_{\rm d}(z,\mathbf{u})\,\mathbf{u}\cdot\mathbf{e}_z\,d\Omega=0\quad\text{for}\quad z=0 \tag{D.4}$$

where the angular integration is over directions satisfying $\mathbf{u}\cdot\mathbf{e}_z>0$. Following the same steps as in section 5.6 , we find that (Eq. 5.18)

$$\frac{v_E}{2}\,U(z=0)+q(z=0)=0\;.$$

Making use of the expression of the energy current Eq. (D.3), we obtain the boundary condition at the interface $z = 0$ in the presence of an incident collimated intensity:

$$U(z=0) - \frac{2}{3}\frac{1}{\mu_a + \mu_s'}\frac{dU}{dz}(z=0) + \frac{2\mu_s\, g}{v_E(\mu_a + \mu_s')}\, I_{\rm b}(z=0) = 0\ . \tag{D.5}$$

Note that in the absence of the collimated source term $I_{\rm b}(z=0) = 0$, we recover the result derived in section 5.6.

Finally, let us note that we have dealt with the simple case of index matched media. In the presence of a refractive index contrast, the boundary condition has to be modified. The extrapolation distance z_0 takes a different value, that accounts for internal reflection of the diffuse intensity at the interface. For a detailed study and the derivation of practical expressions, see Refs. [18, 19, 13] and [1] (chap. 25).

Diffuse transmission factor

In this section we calculate the power transmitted through a scattering layer with thickness $L \gg \ell_t$ illuminated by a plane wave, in the framework of the diffusion approximation (see Fig. D.2). We assume a non absorbing medium ($\mu_a = 0$).

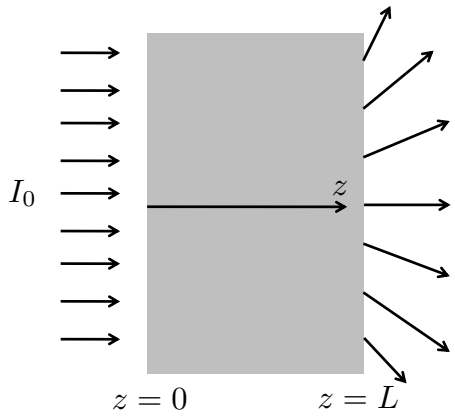

Figure D.2: Diffuse transmission through a slab with thickness L. The interface $z = 0$ is illuminated by a plane wave with intensity I_0 (W.m^{-2}).

The energy density $U(z)$ in the medium satisfies (Eq. D.2 with $\mu_a = 0$)

$$\frac{d^2\, U}{dz^2} = -\frac{3\mu_s^2}{v_E}\, I_{\rm b}(z) \tag{D.6}$$

where $\mu_s' = \mu_s(1-g) = 1/\ell_t$ and $I_{\rm b}(z) = I_0 \exp(-\mu_s z)$.

The general solution is the sum of the solution to $d^2\,U/dz^2 = 0$ and a particular solution of Eq. (D.6). Using $U_p(z) = (-3I_0/v_E)\exp(-\mu_s z)$ as a particular solution, we have

$$U(z) = C_1 z + C_2 - \frac{3I_0}{v_E}\,\exp(-\mu_s z) \tag{D.7}$$

where C_1 and C_2 are two constants to be determined. To proceed, we use the boundary conditions at the interfaces. At the interface $z = 0$, the boundary condition is (see Eq. D.5)

$$U(z=0) - \frac{2}{3}\frac{1}{\mu_a + \mu_s'}\,\frac{dU}{dz}(z=0) + \frac{2\mu_s\,g}{v_E(\mu_a+\mu_s')}\,I_{\rm b}(z=0) = 0\;. \tag{D.8}$$

At the interface $z = L$, it is easy to see that using the procedure described in Appendix B we obtain

$$U(z=L) + \frac{2}{3}\frac{1}{\mu_a + \mu_s'}\,\frac{dU}{dz}(z=L) - \frac{2\mu_s\,g}{v_E(\mu_a+\mu_s')}\,I_{\rm b}(z=L) = 0\;. \tag{D.9}$$

Inserting Eq. (D.7) into the above boundary conditions, the expressions of the constants C_1 and C_2 are readily obtained. For $z \gg \ell_s$, all terms proportionnal to $\exp(-\mu_s z)$ can be neglected, and the expression of the energy density inside the medium becomes

$$U(z) = \frac{5I_0}{v_E}\left[\frac{L + z_0 - z}{L + 2z_0}\right] \tag{D.10}$$

where $z_0 = (2/3)\,\ell_t$.

The transmitted flux (unit W.m^{-2}) is

$$\phi = -D\,\frac{dU}{dz}(z=L) \tag{D.11}$$

where $D = (1/3)v_E\ell_t$ is the diffusion constant. We can define a transmission coefficient $T = \phi/I_0$, which can be deduced from Eqs. (D.10) and (D.11):

$$T = \frac{5}{3}\frac{\ell_t}{L + 2z_0} \simeq \frac{5}{3}\frac{\ell_t}{L} \tag{D.12}$$

since we have assumed $L \gg \ell_t$. The scaling of the transmission coefficient with $1/L$ is a feature of diffusive transport. For example, Ohm's law gives an electrical conductance proportionnal to $1/L$ with L the length of the conductor.

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