ESPCI PARIS | PSL

3ème année

Waves in Complex Media

Lecture notes

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Foreword

These lecture notes of the course on *Waves in Complex Media* given at ESPCI Paris-PSL summarize the basic concepts and the technical developments. They do not include the many examples and applications that are discussed all along the lecture.

The lecture assumes that the reader has basic knowledge in wave physics (electromagnetic waves, acoustics, optics, propagation and diffraction).

As additional reading, the reader wishing to deepen the course can rely on the following reference, which naturally completes the presentation:

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 is negligible.

1.1 Formal description of a scattering problem

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

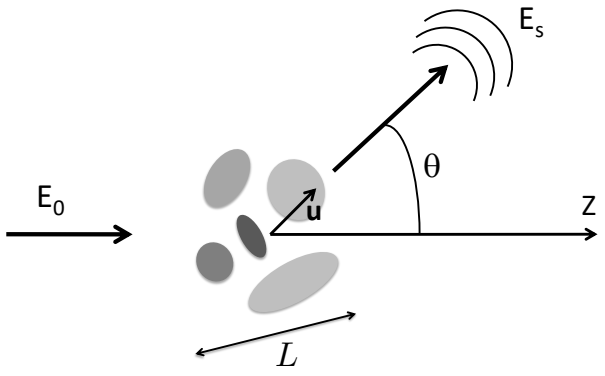

Figure 1.1: Geometry of the scattering problem. A heterogeneous material (scattering medium) is enclosed in a volume with typical size L.

The scattering medium fills a finite volume (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. 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).[2] 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, with L the size of the scattering volume. Using this expansion in Eq. (1.8), we find that the scattered field in the far zone takes the form

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

[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 [2].

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

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

When the incident field is a plane wave with complex amplitude $E_0(\mathbf{r}) = A_0 \exp(i\mathbf{k}_{inc}\cdot\mathbf{r})$, where $\mathbf{k}_{inc}$ is the incident wavevector satisfying $|\mathbf{k}_{inc}| = k_0$, 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 V enclosed by a surface S. Using Eq. (1.1) in volume 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 so that for a plane wave we simply have $|\mathbf{J}| = |E|^2$. 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

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

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

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

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

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

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

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

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

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

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

Integrating over volume V, and noting that

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

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

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

Since $E_s = E - E_0$, the integrand can be written $\mathrm{Im}[VE_s^*E] = \mathrm{Im}V|E|^2 - \mathrm{Im}[VE_0^*E]$. 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

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

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.

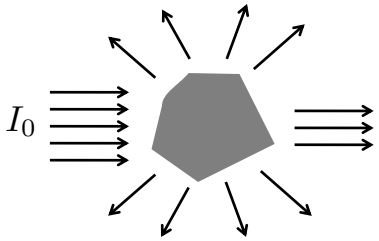

Figure 1.2: Schematic representation of extinction of an incident beam by a scattering object.

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 $E_0(\mathbf{r}) = A_0 \exp(ik_0 \mathbf{u}_{inc} \cdot \mathbf{r})$, with $\mathbf{u}_{inc}$ the unit vector defining the direction of incidence, Eq. (1.27) becomes

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

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]

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

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.

1.3.1 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.

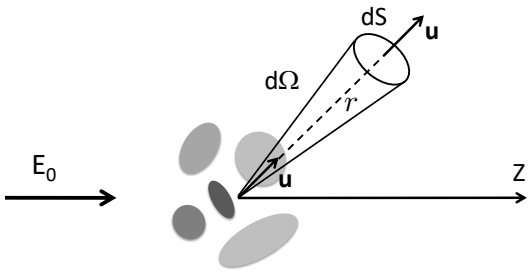

Figure 1.3: Geometry used in the defintion of the power scattered in a given direction. The surface dS supported by the solid angle $d\Omega$ at a distance r is $dS = r^2\, d\Omega$. The flux scattered in the solid angle $d\Omega$ coincides with the flux through dS.

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:[5]

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

[5]Integration over the solid angle Ω is equivalent to integration over all directions of the unit vector $\mathbf{u}$.

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).

1.3.2 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 $\sigma_a = 0$.

1.3.3 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.

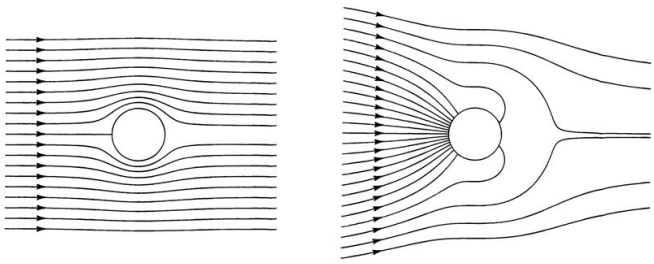

Figure 1.4: Flux lines of an optical plane wave interacting with a silver nanoparticle. The oscillation of free electrons in the metal leads to a resonance (plasmon resonance) that enhances extinction. Left: off resonance. Right: on resonance. The right figure shows a situation in which the extinction cross section is larger than the geometrical cross section. Adapted from Ref. [3].

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 the full vector formalism is used). 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 same lines as 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 current density $\mathbf{j}_{ext}$. In the absence of any other object, the electric field produced by this source is the incident field $\mathbf{E}_0$.

In the presence of the particle (scatterer), the total field is

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

where $\mathbf{E}_s$ denotes the scattered field, that has to be understood as the field radiated by the induced current (or polarization) in the particle. This statement simply reflects the superposition theorem.[1] Solving the scattering problem amounts to calculating $\mathbf{E}_s(\mathbf{r})$ in order to deduce, for example, the scattered or absorbed power.

[1] In a standard scattering problem it is assumed that the source of the incident field $\mathbf{j}_{ext}$ is not modified by the presence of the scatterer.

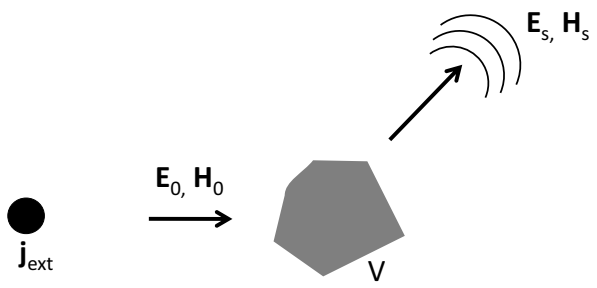

Figure 2.1: Scattering of an incident electromagnetic wave by a particle with volume V.

Describing the particle 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

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

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

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 $\mathbf{G}_0$, 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 the outgoing wave condition when $|\mathbf{r} - \mathbf{r}'| \to \infty$ (this condition means that the Green function behaves as an outgoing spherical wave when $|\mathbf{r} - \mathbf{r}'| \to \infty$). 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[2]

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

[2]Here we favor an intuitive approach, with the Green's function seen as a linear impulse response. Equation (2.5) can be established formally using the vector form of the second Green identity, see for example [1] (chap. 37) or [2].

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

Consider the particular case of a point electric dipole source located at a point $\mathbf{r}_0$. The associated current density is $\mathbf{j}_{ext} = -i\omega\mathbf{p}\delta(\mathbf{r}-\mathbf{r}_0)$, 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 $\mathbf{G}_0(\mathbf{r},\mathbf{r}')$ can be understood as the electric field radiated at point $\mathbf{r}$ in free space by an elementary point source (electric dipole) located at point $\mathbf{r}'$. For electromagnetic waves, the Green function is a second-rank tensor (that can be represented as a 3×3 matrix). The above relation can be rewritten as

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \mu_0\,\omega^2 \begin{pmatrix} G_{xx} & G_{xy} & G_{xz} \\ G_{yx} & G_{yy} & G_{yz} \\ G_{zx} & G_{zy} & G_{zz} \end{pmatrix} \begin{pmatrix} p_x \\ p_y \\ p_z \end{pmatrix}.$$

Equation (2.6) 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 [4]:

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

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 $[\mathbf{u}'\otimes\mathbf{u}']_{ij} = u_i'u_j'$ 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 scattered field. In the far-field limit, the Green's function can be simplified into

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

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 field radiated by the dipole is a spherical wave, corrected by a phase term that accounts for the shift in position of the dipole with respect to the origin of coordinates. The tensor term is simply the projection on the direction perpendicular to $\mathbf{u}$ (remember that the electric field has to be transverse in the far field). This is the main difference with the scalar Green's function in Eq. (1.11).

2.1.3 Integral equation

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

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

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 from Eq. (2.10) by adding the incident field:

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

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 with complex amplitude $\mathbf{E}_0(\mathbf{r}) = E_0\,\mathbf{e}_0\,\exp(i\mathbf{k}_{inc}\cdot\mathbf{r})$, with $\mathbf{e}_0$ a unit vector defining the direction of polarization. In this case, it is useful to introduce the normalized scattering matrix $\mathbf{S}(\mathbf{u})$ 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. In practice, the scattering matrix for polarized light can be described in terms of Stokes vectors, a formalism that is not used in this lecture but that is briefly reviewed in Appendix A.

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[3], 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.[4] 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

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

[3] 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 $(\omega/2)\,\mathrm{Im}(\mathbf{P}\cdot\mathbf{E}^*)$.

[4] 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[5]. 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[6]

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

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:

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

[5] 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].

[6] 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 these 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 $(k_0 R)^3$, and tends to zero when $k_0 R \ll 1$. 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 [4], 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 [4]. 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. [3]), 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.

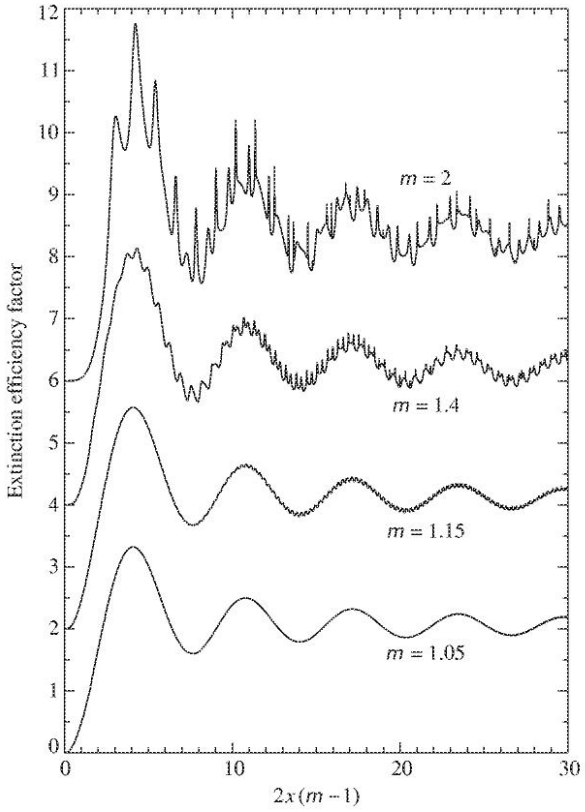

Figure 2.2: Extinction efficiency $Q_e = \sigma_e/(\pi R^2)$ of a spherical particle with radius R and refractive index m. The parameter $x = 2\pi R/\lambda$, where λ is the incident wavelength, is the size parameter. Adapted from [8].

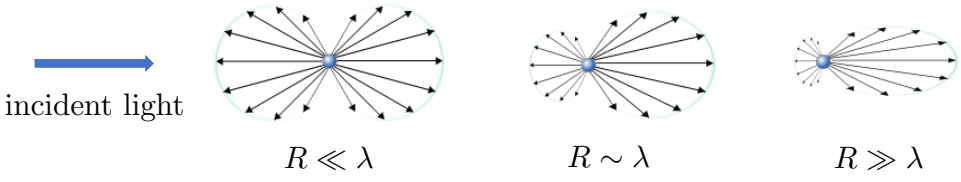

Figure 2.3: Scattering diagrams for spherical particles with different sizes. Large particles produce a strong forward scattering. Adpated from Wikipedia.

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 (particles). We define the different scattering regimes, and introduce the statistical approach that will be used all along the lecture.

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 the field obeys the Lippmann-Schwinger equation, which reads as

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

with E_0 the complex amplitude of the incident wave.

In the following it will be convenient to use an operator notation, that allows us to rewrite Eq. (3.1) in the compact form

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

In this notation, E stands for a "state vector" $|E\rangle$ (as in quantum mechanics), and G_0 and V are operators such that $G_0 : |f\rangle \rightarrow \int G_0(\mathbf{r},\mathbf{r}')f(\mathbf{r}')d^3r'$ and $V : |f\rangle \rightarrow V(\mathbf{r})f(\mathbf{r})$. Upon iterating Eq. (3.2) we obtain

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

which is known as the Born series. Limiting the expansion to $E = E_0 + G_0\, V\, E_0$ corresponds to the Born approximation, and defines the regime of single scattering. The other terms in the expansion correspond to higher orders of multiple scattering.

In order to derive an expression of the scattered field in which scattering between different particles becomes explicit, it will prove useful to introduce the transition operator, or T matrix, defined as

$$T = V + VG_0V + VG_0VG_0V + ... \tag{3.4}$$

which in real space is also

$$\begin{aligned} T(\mathbf{r}_1, \mathbf{r}_2) &= V(\mathbf{r}_1)\,\delta(\mathbf{r}_1 - \mathbf{r}_2) + V(\mathbf{r}_1)\,G_0(\mathbf{r}_1, \mathbf{r}_2)\,V(\mathbf{r}_2) \\ &+ \int V(\mathbf{r}_1)\,G_0(\mathbf{r}_1, \mathbf{r}')\,V(\mathbf{r}')\,G_0(\mathbf{r}', \mathbf{r}_2)\,V(\mathbf{r}_2)\,d^3r' + ... \end{aligned} \tag{3.5}$$

A summation of the geometric series gives

$$T = V(1 - G_0V)^{-1} \tag{3.6}$$

which is a formal expression of T in terms of V.

The Born series (3.3) can be written

$$E = E_0 + G_0\,(V + V\,G_0\,V + V\,G_0\,V\,G_0\,V + ...)\,E_0 \tag{3.7}$$

in which the series in the parenthesis is recognized as the T matrix. This allows us to rewrite the Lippmann-Schwinger equation (3.2) in the form

$$E = E_0 + G_0\,T\,E_0\,. \tag{3.8}$$

We note that the field on the right is the incident field E_0. The 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 will now see.

3.1.2 Set of discrete scatterers

The potential V and the T matrix describe the scattering medium as a whole. For an ensemble of discrete scatterers, the potential can be written

$$V(\mathbf{r}) = \sum_j V_j(\mathbf{r}) \tag{3.9}$$

with $V_j(\mathbf{r})$ the potential due to the scatterer located at position $\mathbf{r}_j$. For light, assuming a set of identical scatterers with dielectric function ϵ, we would have

$$V_j(\mathbf{r}) = k_0^2(\epsilon - 1)\,\Theta(\mathbf{r} - \mathbf{r}_j) \tag{3.10}$$

where $\Theta(\mathbf{r}-\mathbf{r}_j)$ is the function such that $\Theta(\mathbf{r}-\mathbf{r}_j) = 1$ if $\mathbf{r}$ is inside the particle and $\Theta(\mathbf{r}-\mathbf{r}_j) = 0$ otherwise.

Using Eq. (3.9) and the defintion of the T matrix (3.5), we can write

$$\begin{aligned} T(\mathbf{r}_1,\mathbf{r}_2) &= \sum_j V_j(\mathbf{r}_1)\,\delta(\mathbf{r}_1-\mathbf{r}_2) + \sum_{j,k} V_k(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}_2)\,V_j(\mathbf{r}_2) \\ &+ \sum_{j,k,l}\int V_l(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}')\,V_k(\mathbf{r}')\,G_0(\mathbf{r}',\mathbf{r}_2)\,V_j(\mathbf{r}_2)\,d^3r' + \ldots \end{aligned} \tag{3.11}$$

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

$$\begin{aligned} t_j(\mathbf{r}_1,\mathbf{r}_2) &= V_j(\mathbf{r}_1)\,\delta(\mathbf{r}_1-\mathbf{r}_2) + V_j(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}_2)\,V_j(\mathbf{r}_2) \\ &+ \int V_j(\mathbf{r}_1)\,G_0(\mathbf{r}_1,\mathbf{r}')\,V_j(\mathbf{r}')\,G_0(\mathbf{r}',\mathbf{r}_2)\,V_j(\mathbf{r}_2)\,d^3r' + \ldots \end{aligned} \tag{3.12}$$

This operator describes the scattering properties of a single scatterer, and is built in the same way as the global T matrix. Its use will allow us to separate the scattering process occuring inside a single scatterer, and the scattering process occuring between different scatterers. Using Eq. (3.12), we see that the global T matrix can be written in terms of the T matrix of the individual scatterers:

$$T = \sum_j t_j + \sum_{j\neq k} t_j\,G_0\,t_k + \sum_{j\neq k,k\neq l} t_j\,G_0\,t_k\,G_0\,t_l + \ldots \tag{3.13}$$

One can check that by inserting (3.12) into (3.13) all terms in Eq. (3.11) are recovered. We obtain the final expression of the field by inserting Eq. (3.13) into Eq. (3.8):

$$E = E_0 + \sum_j G_0\,t_j\,E_0 + \sum_{j\neq k} G_0\,t_j\,G_0\,t_k\,E_0 + \sum_{j\neq k,k\neq l} G_0\,t_j\,G_0\,t_k\,G_0\,t_l\,E_0 + \ldots \tag{3.14}$$

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 sequences. 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.

Particular case: T matrix of a small scatterer

We have seen in section 2.3 that for light, and for a scatterer much smaller than the wavelength treated in the electric dipole approximation, the scattering properties are determined by the polarizability $\alpha(\omega)$. Using Eq. (2.40) in the scalar model, we write that the field scattered by a single scatterer at position $\mathbf{r}_j$ is

$$E_s(\mathbf{r}) = k_0^2 G_0(\mathbf{r},\mathbf{r}_j)\alpha(\omega)E_0(\mathbf{r}_j)\,. \tag{3.15}$$

In terms of the T matrix we can also write

$$E_s(\mathbf{r}) = \int G_0(\mathbf{r}, \mathbf{r}_1) t_j(\mathbf{r}_1, \mathbf{r}_2) E_0(\mathbf{r}_2) d^3 r_1 d^3 r_2 \,. \tag{3.16}$$

Since the above expressions must coincide, we find that the T matrix of a single small scatterer is $t_j(\mathbf{r}_1, \mathbf{r}_2) = k_0^2 \, \alpha(\omega) \, \delta(\mathbf{r}_1 - \mathbf{r}_j) \delta(\mathbf{r}_2 - \mathbf{r}_j)$.

3.2 Field propagator and scattering sequences

Consider the canonical slab geometry shown in Fig. 3.1. The transmitted field $E(\mathbf{r}_b)$ at a point

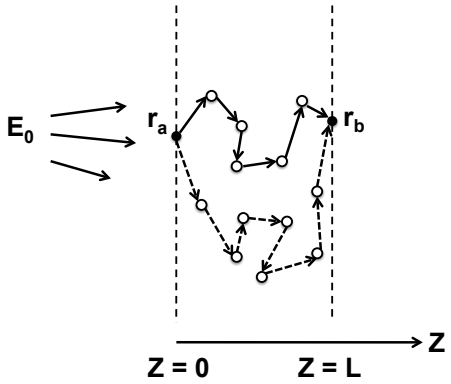

Figure 3.1: Schematic representation of scattering sequences in a slab geometry. White circles stand for scattering events (scatterers). Black point are entry and exit points on the slab surfaces (that do not necessarily coincide with scattering events).

$\mathbf{r}_b$ 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

$$E(\mathbf{r}_b) = \int_{z=0} h(\mathbf{r}_b, \mathbf{r}_a) \, E_0(\mathbf{r}_a) \, d^2 \rho_a \tag{3.17}$$

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 surface $z = 0$.

We will now see that Eq. (3.14) allows us to write the amplitude propagator in the form of a summation over scattering sequences:

$$h(\mathbf{r}_b, \mathbf{r}_a) = \sum_{n=0}^{\infty} \sum_{\mathcal{S}_n = \{\mathbf{r}_1, \mathbf{r}_2 \ldots \mathbf{r}_n\}} A_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a) \, \exp[i \phi_{\mathcal{S}_n}(\mathbf{r}_b, \mathbf{r}_a)] \tag{3.18}$$

In this expression, we define a scattering sequence $\mathcal{S}_n$ with n scattering events by the positions $\{\mathbf{r}_1, \mathbf{r}_2 ... \mathbf{r}_n\}$ of the successive scattering events (remember that $\mathbf{r}_a$ and $\mathbf{r}_b$ do not necessarily coincide with scattering events). The summation includes all scattering sequences with n scattering events, and runs for $n = 0 \rightarrow \infty$ ($n = 0$ 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

$$h(\mathbf{r}_b, \mathbf{r}_a) = \sum_{\mathcal{S}_{ab}} A_{\mathcal{S}_{ab}} \exp(i\phi_{\mathcal{S}_{ab}}) \tag{3.19}$$

where $\mathcal{S}_{ab}$ stands for any sequence connecting $\mathbf{r}_a$ to $\mathbf{r}_b$. Two scattering sequences are represented schematically in Fig. 3.1.

To convince ourselves that expression (3.18) can be deduced from Eq. (3.14), let us write explicitly the single scattering term in Eq. (3.14) at point $\mathbf{r}_b$:

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

$$\int \left[\sum_j \int G_0(\mathbf{r}_b, \mathbf{r}_1)\, t_j(\mathbf{r}_1, \mathbf{r}_2)\, h_0(\mathbf{r}_2, \mathbf{r}_a) d^3r_1 d^3r_2 \right] E_0(\mathbf{r}_a) d^2\rho_a \tag{3.20}$$

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. (3.14) can be written exlicitly the same way. By comparing to Eq. (3.17), we deduce

$$\begin{aligned} h &= h_0 + \sum_j G_0\, t_j\, h_0 + \sum_{j,k} G_0\, t_k\, G_0\, t_j\, h_0 \\ &+ \sum_{j \neq k, k \neq l} G_0\, t_l\, G_0\, t_k\, G_0\, t_j\, h_0 + \ldots \end{aligned} \tag{3.21}$$

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. Therefore, expression (3.21) is exactly of the form given by Eq. (3.18). Representations in the form of scattering sequences are very useful, in particular in the study of speckle.

[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.

3.3 Statistical approach

In a disordered medium, a precise description of the detailed microstructure is out of reach. Most of the time, a microscopic description would even 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), that depend on a few statistical parameters characterizing the disordered medium (for example the average number of scatterers per unit volume). Instead of describing precisely a particular realization of a disordered medium, and then performing some statistical analysis, we can use a statistical approach in the first place, and deduce the statistical properties of the observables without solving the full microscopic problem on a specific realization. For example, this approach will allow us to find the expression of the averaged transmissivity of a glass of milk, without solving the equations of light scattering in a frozen configuration of colloidal particles in suspension in water. This is actually the spirit of any approach in statistical physics. To proceed, we consider conceptually an ensemble of realizations of the disordered medium, and perform an ensemble averaging denoted by $\langle ... \rangle$. Once this statistical point of view has been adopted, we often speak of wave scattering in random media (although the randomness results more from the description of the problem than from the medium itself).

3.3.1 Average field and fluctuations

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

$$E = \langle E \rangle + \delta E \quad \text{with} \quad \langle \delta E \rangle = 0 \,. \tag{3.22}$$

The first term 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

$$E = E_0 + \langle E_s \rangle + \delta E_s \tag{3.23}$$

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

3.3.2 Average intensity

The average intensity is $\langle I \rangle = \langle |E|^2 \rangle$. Using Eq. (3.22), we immediatly see that

$$\langle I \rangle = |\langle E \rangle|^2 + \langle |\delta E|^2 \rangle \,. \tag{3.24}$$

The first term is the power carried by the average field. It represents the ballistic component I_b of the average intensity. 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. 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 $I_\mathrm{d} = \langle |\delta E|^2 \rangle$.

It is instructive to rewrite the ballistic intensity in the form

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

in which the last term describes the interference between the average scattered field and the incident field. Intuitively, extinction by scattering and absorption must impose $I_\mathrm{b} < |E_0|^2$, 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 we discussed in chapter 1.

Formally, it is possible to derive the equations satisfied by the average field (Dyson equation) and by the average intensity (Bethe-Salpeter equation). A detailed presentation of this multiple scattering theory can be found in review articles and textbooks [1, 9, 10, 11, 12]. These equations remain very difficult to handle, and their use for practical calculations of the average intensity usually requires approximations that lead to transport equations that can also be derived phenomenologically. In this lecture we favor the phenomenological approach to describe the ballistic and diffuse intensity. The treatment of the ballistic intensity is the subject of the next section, while the treatment of the diffuse intensity is the subject of the next two chapters.

3.4 Ballistic intensity

Let us consider a scattering medium in the slab geometry in Fig. 3.2, illuminated by a plane wave with complex amplitude E_0. From Eq. (3.8), the average field is

$$\langle E \rangle = E_0 + G_0 \langle T \rangle E_0 \tag{3.25}$$

since the incident field 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 T matrix is homogeneous and isotropic. From the equation above, we can conclude that the average field sees an effective homogeneous and isotropic medium. This means that if the incident field is a plane wave (in practice a collimated beam), then the average field is also a plane wave that is partially

reflected and transmitted by a slab of an effective homogeneous and isotropic material. Due to scattering and absorption, the average field is also attenuated and the effective medium is lossy (note that the averaged field is attenuated by scattering even in the absence of absorption).

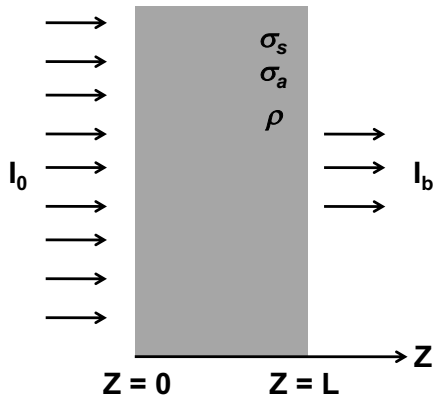

Figure 3.2: Attenuation of a collimated beam (plane wave) by a slab with thickness L filled with a statistically uniform scattering and absorbing material, made of identical particles with number density ρ, and scattering and absorption cross sections σ_s and σ_a.

In order to find the attenuation of the ballistic intensity $I_\mathrm{b} = |\langle E \rangle|^2$ transmitted through a slab with thickness L, 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 $z + dz$:

$$I_\mathrm{b}(z + dz)\, S - I_\mathrm{b}(z)\, S = -(\rho\, S\, dz)\, (\sigma_a + \sigma_s)\, I_\mathrm{b}(z) = -(\rho\, S\, dz)\, \sigma_e\, I_\mathrm{b}(z)\ . \tag{3.26}$$

This expression describes the extinction of the ballistic intensity between z and $z + dz$, due to the $\rho\, S\, dz$ particules located in the volume.[2] We deduce the equation satisfied by $I_\mathrm{b}(z)$:

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

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

$$I_\mathrm{b}(z) = I_\mathrm{b}(z = 0)\, \exp(-\rho\, \sigma_e\, L)\ . \tag{3.27}$$

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 (3.27) is actually the general form of the Beer-Lambert law.

[2] We assume that each particle scatters as if it were alone in the medium. This is known as the independent scattering approximation, valid in diluted media.

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

$$I_{\rm b}(z) = I_{\rm b}(z=0)\,\exp(-L/\ell_e)\,. \tag{3.28}$$

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

$$\frac{1}{\ell_e} = \frac{1}{\ell_s} + \frac{1}{\ell_a}\,. \tag{3.29}$$

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

Finally, 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 $\mu_e = \rho\sigma_e$, the scattering coefficient $\mu_s = \rho\sigma_s$, and the absorption coefficient $\mu_a = \rho\sigma_a$ (these coefficients are widely used for example in biomedical optics).

3.5 Transport 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). Describing the diffuse intensity is much more involved, and will be the objective of the next two chapters. We will use an approach similar to that used for the transport of particles. To introduce this analogy, we show in this section that the Beer-Lambert law is consistent with a point of view borrowed to the kinetic theory of classical transport.

3.5.1 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. (3.26) in the form

$$I_{\rm b}(z+dz) \;= I_{\rm b}(z) - \frac{dz}{\ell_s}\,I_{\rm b}(z)\,. \tag{3.30}$$

Seeing the intensity as a flux of particles (we will use the term "photons" for convenience, although they have to be considered as classical particles), 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 in the medium, and propagting along a direction that we choose as Oz. The probability for this photon to be scattered *for the first time* after a distance z is

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

where we introduce $P(z)$ in the left-hand side as the probability density. The average distance before the first 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 before the next scattering event for a photon taken at random (this is equivalent to stating that ℓ_s is the average distance between two successive scattering events). The name "mean free path" given to ℓ_s is now clear.

3.5.2 Single and multiple scattering

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

- $L \ll \ell_s$: Ballistic regime (the wave goes through without being scattered)
- $L \simeq \ell_s$: Single scattering regime
- $L \gg \ell_s$: Multiple scattering regime.

In the multiple scattering regime, the ballistic intensity is completely extinguished and energy transport only occurs through the diffuse intensity. At large scales, we will see that the intensity transport obeys a diffusion law.

3.5.3 Localization

In the regime $L \gg \ell_s$ and $\ell_s \simeq \lambda$, with λ the wavelength, a substantial deviation from the diffusion law is expected due to the phenomenon of Anderson localization. The interested

reader could refer to [13] for an introduction to the topic and [9] for a more advanced treatment. The localization regime will not be discussed in this lecture, and we will assume that the condition $\ell_s \gg \lambda$ is satisfied.

3.5.4 Homogenization

It is also interesting to briefly address the regime of homogenization, that 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. 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 will only discuss an example based on scaling laws.

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.

3.6 Diffuse intensity: Towards a transport equation

With reference to the slab geometry in Fig. 3.1, and using Eq. (3.17), the average intensity in the output plane can be written

$$\langle I(\mathbf{r}_b)\rangle = \int_{z=0} \langle h(\mathbf{r}_b, \mathbf{r}_a) h^*(\mathbf{r}_b, \mathbf{r}'_a)\rangle\, E_0(\mathbf{r}_a) E_0^*(\mathbf{r}'_a)\, d^2\rho_a d^2\rho'_a \,. \tag{3.31}$$

The ballistic intensity can be written

$$I_{\rm b}(\mathbf{r}_b) = \int_{z=0} \langle h(\mathbf{r}_b,\mathbf{r}_a)\rangle \, \langle h^*(\mathbf{r}_b,\mathbf{r}'_a)\rangle \, E_0(\mathbf{r}_a) E_0^*(\mathbf{r}'_a) \, d^2\rho_a d^2\rho'_a \tag{3.32}$$

showing that the diffuse intensity $I_{\rm d} = \langle I\rangle - I_{\rm b}$ is driven by the correlator $\langle h(\mathbf{r}_b,\mathbf{r}_a)h^*(\mathbf{r}_b,\mathbf{r}'_a)\rangle - \langle h(\mathbf{r}_b,\mathbf{r}_a)\rangle\langle h^*(\mathbf{r}_b,\mathbf{r}'_a)\rangle$. Note that at large scales ($L \gg \ell_s$), the ballistic intensity is exponentially small and we can assume $\langle I\rangle \simeq I_{\rm d}$.

The key point in evaluating $\langle I(\mathbf{r}_b)\rangle$ is to compute the correlator $\langle h(\mathbf{r}_b,\mathbf{r}_a)h^*(\mathbf{r}_b,\mathbf{r}'_a)\rangle$. Inserting the expansion (3.21) of $h(\mathbf{r}_b,\mathbf{r}_a)$ in scattering sequences, and trying to perform the averaging over the positions of the scatterers, we immediately understand that $\langle h(\mathbf{r}_b,\mathbf{r}_a)h^*(\mathbf{r}_b,\mathbf{r}'_a)\rangle$ is a very complex object. Actually, there is no hope to average by hand, and multiple scattering theory is a framework developed to handle such an averaging process [1, 9, 11, 12]. 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 computation of quadratic quantities, the most general object is the field correlation function $\langle E(\mathbf{r}_b)E^*(\mathbf{r}'_b)\rangle$ (when $\mathbf{r}_b = \mathbf{r}'_B$ this correlation function coincides with the average intensity $\langle I(\mathbf{r}_b)\rangle$). 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 Fig. 3.1. 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. Indeed, even two scattering sequences differing by only one scattering event have a difference in optical path on the order of $\ell_s \gg \lambda$. The interference term between the fields scattered along the two sequences will average to zero due to their large phase difference. As a result, only the contributions resulting from two fields E and E^* following the same scattering sequences will contribute to $\langle E(\mathbf{r}_b)E^*(\mathbf{r}'_b)\rangle$. Keeping only these contributions is known as the ladder approximation. In the formal multiple scattering theory, this approximation emerges from a first-order perturbative expression of $\langle E(\mathbf{r}_b)E^*(\mathbf{r}'_b)\rangle$ in terms of the small parameter $1/(k_0\ell_s)$, with $k_0 = 2\pi/\lambda$ [1, 9, 11, 12].[3]

In the regime $k_0\ell_s \gg 1$, and at large scale $L \gg \ell_s$, the ladder approximation allows us to write

$$\langle h(\mathbf{r}_b,\mathbf{r}_a)h^*(\mathbf{r}'_b,\mathbf{r}'_a)\rangle \simeq P(\mathbf{r}_b,\mathbf{r}_a)\,\delta(\mathbf{r}_b-\mathbf{r}'_b)\,\delta(\mathbf{r}_a-\mathbf{r}'_a) \tag{3.33}$$

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 $\mathbf{r}_b$, as represented in Fig. 3.3. Again, Eq. (3.33) can be justified rigorously in the framework of multiple scattering theory. Also keep in mind that the delta functions make sense only for the computation of field correlations, or intensities, at scales larger than the scattering mean free path ℓ_s (macroscopic description).

[3] To get an order of magnitude, we can think of the propagation of near infrared light in biological tissues. The wavelength is $\lambda \simeq 1$ μm, and the scattering mean free path is $\ell_s \simeq 100$ μm. Therefore $k_0\ell_s \simeq 600$ and $1/(k_0\ell_s) \ll 1$.

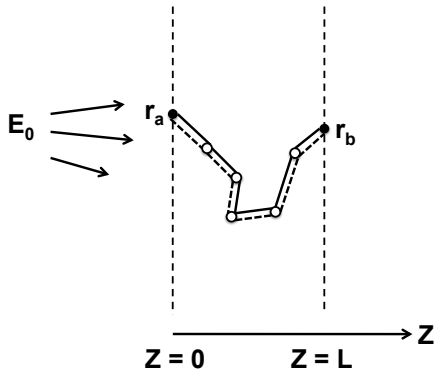

Figure 3.3: Graphical representation of the ladder approximation in a slab geometry. The diffuse intensity is transported through scattering paths involving the same sequence of scatterers for E (solid line) and E^* (dashed line).

In summary, we have ended up with the following picture: in the regime $k_0\ell_s \gg 1$, the diffuse intensity can be understood as the sum of *intensity* contributions along 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, based on the radiative transfer equation, and on the diffusion approximation.

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 developed later to describe the transport of neutrons in nuclear reactors [15]. A derivation of the RTE starting from the wave equation is available, and relies on the ladder approximation briefly discussed at the end of chapter 3 (see for example [1] (chap. 16 and 17), [10] or [11]).

4.1 Specific intensity

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

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

where $d\Omega$ means an integration over the solid angle, or equivalently over the direction defined by the unit vector $\mathbf{u}$ (we have $d\Omega = \sin\theta\, d\theta\, d\phi$ in spherical coordinates). In this expression, $I(\mathbf{r},\mathbf{u},\omega,t)$ is the specific intensity that represents the power per unit surface flowing in direction $\mathbf{u}$ at point $\mathbf{r}$ and time t (the unit of the specific intensity is W.m^{-2}.sr^{-1}).[1] Here we assume quasi-monochromatic waves, with a central frequency ω and a slowly varying envelope in time. **For the sake of simplicity in the following, we will omit the variable ω in the notations, and write for example $I(\mathbf{r},\mathbf{u},t)$ for the specific intensity at a given frequency ω.**

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

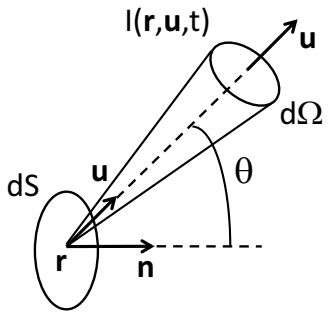

Figure 4.1: Geometry used for the definition of the specific intensity.

From the specific intensity we can define the energy current by

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

It coincides (up to an arbitrary prefactor) with the energy current $\mathbf{J}$ defined in chapter 1 (Eq. 1.17). The flux of $\mathbf{q}$ through a surface is the gobal energy flux (integrated over all directions), flowing through this surface (unit W).

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 $d\Omega$.[2] It is easy to see that in this case $\mathbf{q}(\mathbf{r},t) = v_E\mathbf{u}_0\,U(\mathbf{r},t)$, which gives a clear meaning to the energy velocity.[3] For an isotropic radiation, the specific intensity is independent on $\mathbf{u}$, and we have $I(\mathbf{r},t) = (v_E/4\pi)\,U(\mathbf{r},t)$ and $\mathbf{q}(\mathbf{r},t) = 0$.

4.2 Loss and gain processes

We will now describe the evolution of the specific intensity in a scattering and absorbing medium. The latter is considered at the macroscopic scale, with scattering and absorption

[2] We have $\delta(\mathbf{u}-\mathbf{u}_0) = \delta(\theta-\theta_0)\,\delta(\phi-\phi_0)/|\sin\theta_0|$.

[3] Note the analogy with the relation $\mathbf{j} = \rho\mathbf{v}$ between the current density, the charge density and the charge velocity in electrodynamics.

processes modelled using continuous parameters (such as the absorption and scattering coefficients). These parameters have to be understood as averaged over a small volume (a point at the macroscopic scale) large enough to contain many particles. We will assume that macroscopically the medium is homogeneous and isotropic.

4.2.1 Absorption

Consider the energy flux propagating along a direction $\mathbf{u}$. After propagating over a distance ds in the medium, the decrease of the specific intensity due to absorption can be written

$$\mathrm{d}I_a(\mathbf{r},\mathbf{u},t) = -\mu_a\, I(\mathbf{r},\mathbf{u},t)\, ds \tag{4.4}$$

where μ_a is the absorption coefficient (unit m^{-1}). Its inverse $\ell_a = 1/\mu_a$ is the absorption mean free path (or absorption length). In diluted media made of identical particles, and in the independent scattering regime (no correlations in the positions of different particles), we have $\mu_a = \rho\,\sigma_a$, where ρ is the number density of particles and σ_a their absorption cross section.

4.2.2 Extinction by scattering

The process of scattering also removes some energy from the directional flux, and the decrease of the specific intensity due to scattering can be written in a similar fashion:

$$\mathrm{d}I_s(\mathbf{r},\mathbf{u},t) = -\mu_s\, I(\mathbf{r},\mathbf{u},t)\, ds \tag{4.5}$$

with μ_s the scattering coefficient. Its inverse $\ell_s = 1/\mu_s$ is the scattering mean free path (or scattering length). In the independent scattering regime we also have $\mu_s = \rho\,\sigma_s$ with σ_s the scattering cross section of a single particle.

It is convenient to introduce the extinction coefficient $\mu_e = \mu_a + \mu_s$ as well as its inverse $\ell_e = 1/\mu_e$ known as the extinction mean free path (or extinction length). To characterize the relative weight of scattering and absorption in the extinction process, we can use the albedo

$$a = \frac{\mu_s}{\mu_s + \mu_a} \tag{4.6}$$

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

4.2.3 Gain by scattering

The scattering process distributes the incident energy over all directions. To describe this phenomenon, we introduce the phase function $p(\mathbf{u},\mathbf{u}')$ such that $(\mu_s/4\pi)\,p(\mathbf{u},\mathbf{u}')ds$ is the

fraction of the incident power propagating along direction $\mathbf{u}'$ that is scattered in direction $\mathbf{u}$, after a propagation over a distance ds. With this definition, $p(\mathbf{u},\mathbf{u}')$ is a dimensionless quantity, that can be understood as the probability of scattering from direction $\mathbf{u}'$ to $\mathbf{u}$. Note that with this definition the phase function is normalized as $\int_{4\pi} p(\mathbf{u},\mathbf{u}')\,d\Omega = 4\pi$.

For a diluted medium made of identical particles, and in the independent scattering regime, the phase function is simply

$$p(\mathbf{u},\mathbf{u}') = \frac{4\pi}{\sigma_s}\frac{d\sigma_s}{d\Omega} \tag{4.7}$$

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

Using the phase function, the specific intensity is increased by scattering, after propagating along a distance ds, by an amount

$$\mathrm{d}I_g(\mathbf{r},\mathbf{u},t) = \frac{\mu_s}{4\pi}\int_{4\pi} p(\mathbf{u},\mathbf{u}')\,I(\mathbf{r},\mathbf{u}',t)\,d\Omega'\,ds\ . \tag{4.8}$$

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 in the following**.
- If the phase function is a constant (independent on Θ), we speak of isotropic scattering. The other situations correspond to anisotropic scattering. Some examples of phase functions are given in Appendix B.

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

$$g = \frac{1}{4\pi}\int_{4\pi} \mathbf{u}\cdot\mathbf{u}'\ p(\mathbf{u}\cdot\mathbf{u}')\,d\Omega = \frac{1}{4\pi}\int_{4\pi} \cos\Theta\ p(\cos\Theta)\,d\Omega\ . \tag{4.9}$$

In other words, g is the averaged cosine of the scattering angle. $g = 0$ corresponds to isotropic scattering, and $g \simeq 1$ correspond to strong forward scattering. In a medium made of identical particles, and in the independent scattering regime, 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]

[4] In the presence of 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. This is a regime of dependent scattering.

4.3 Energy balance and RTE

The energy flux propagating along direction $\mathbf{u}$ evolves, upon propagation along a distance ds, according to

$$I(\mathbf{r}+d\mathbf{r},\mathbf{u},t+dt)-I(\mathbf{r},\mathbf{u},t)=\mathrm{d}I_a(\mathbf{r},\mathbf{u},t)+\mathrm{d}I_s(\mathbf{r},\mathbf{u},t)+\mathrm{d}I_g(\mathbf{r},\mathbf{u},t)$$

where $d\mathbf{r}=ds\,\mathbf{u}$, and $dt=ds/v_E$ since the energy propagates with a velocity v_E. This balance accounts for extinction by absorption and scattering, and for gain by scattering. Using the explicit expression of the terms in the right-hand side we obtain

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

The left-hand side can be rewritten as

$$\frac{\partial}{\partial t}I(\mathbf{r},\mathbf{u},t)\,dt+\mathbf{u}\cdot\nabla I(\mathbf{r},\mathbf{u},t)\,ds\,, \tag{4.10}$$

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

$$\frac{1}{v_E}\frac{\partial}{\partial t}I(\mathbf{r},\mathbf{u},t)+\mathbf{u}\cdot\nabla I(\mathbf{r},\mathbf{u},t)=-\mu_e\,I(\mathbf{r},\mathbf{u},t)+\frac{\mu_s}{4\pi}\int_{4\pi}p(\mathbf{u}\cdot\mathbf{u}')\,I(\mathbf{r},\mathbf{u}',\omega,t)\,d\Omega' \tag{4.11}$$

which is our final result. 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.

4.4 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:

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

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 (4.11), 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

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

In the steady-state regime, solving this equation leads to the Beer-Lambert law. Indeed, choosing the Oz axis along the direction $\mathbf{u}_0$, we find $I_{\rm b}(z) = I_{\rm b}(0)\exp(-z/\ell_e)$, and we recover the exponential decay of the ballistic beam with a characteristic length $\ell_e = 1/\mu_e$. 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 real situations. Since diffusion processes are found in many transport phenomena, the diffusion equation allows one to draw interesting analogies, and to put forward universal behaviors in wave transport.

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

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

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, it can be usefuel to split the specific intensity into its ballistic and diffuse components (as in section 4.4). The derivation of the diffusion equation starting from the RTE with a source term accounting for the conversion of ballistic intensity into diffuse intensity (as in Eq. 4.14) is given in Appendix C.

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 direction $\mathbf{u}$ (integration over

the solid angle):

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

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) simplifies into

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

which is a local energy balance.

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

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

which takes the form of a continuity equation describing the conservation of the wave energy.

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 the directions $\mathbf{u}$ (this amounts to taking the first moment of the RTE in terms of the angular variables). We obtain:

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

The integral in the right-hand side can be simplified. Using the anistropy parameter g defined in Eq. (4.9), we can show that:[1]

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

Using (5.6), Eq. (5.5) becomes:

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

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.

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 can enter the regime of multiple scattering. After a propagation distance larger than the transport mean free

[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. 4.9), this contribition is $4\pi g$. We deduce that $\int_{4\pi} \mathbf{u}\, p(\mathbf{u}\cdot\mathbf{u}')\, d\Omega = 4\pi g \mathbf{u}'$.

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 (see Appendix A), and keeping only the first two terms, which is known as the P_1 approximation. Under this approximation, the specific intensity can be written [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:

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

[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_ib_i has to be understood as $\sum_i a_ib_i$.

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

where we have assumed $\mu_a \ll \mu_s(1-g)$, or equivalently $\ell_a \gg \ell_t$, to neglect 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. We finally end up with a relationship between the energy current and the gradient of the energy density:

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

This relation 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.

5.4.2 Diffusion equation

Inserting Eq. (5.12) into Eq. (5.3), we obtain the diffusion equation satisfied by the energy density:

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

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 multiple scattering regime, and 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. Qualitatively, this result also shows that in the diffusive regime the wave transport can be undesrtood as a transport of 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 that reads 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 $1/G$ scaling as 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, and is performed in Appendix D. We obtain

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

The decay of the diffuse transmission in $1/L$ in the diffusive regime explains the high reflectivity $R = 1 - T$ of strongly scattering materials. Indeed, a non absorbing and scattering medium illuminated with white light reflects all wavelengths when $L \gg \ell_t$, 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.

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 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 that a wide class of them, known as fully developed speckles, exhibit universal statistics. In this chapter, we study the statistical distribution of the intensity measured at one point in a speckle pattern.

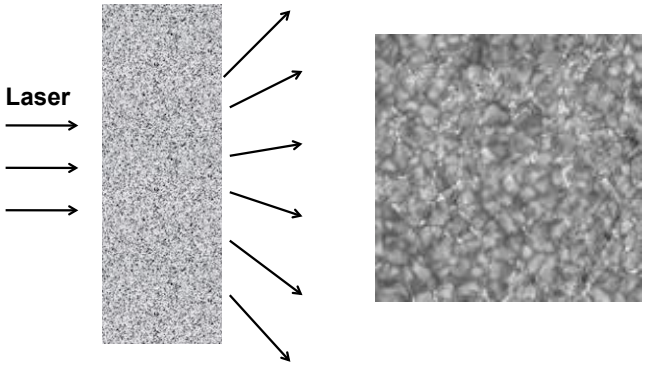

Figure 6.1: Optical speckle pattern generated by illuminating a slab of scattering medium (corresponding to one realization of disorder) with a coherent laser beam.

6.1 Fully developed speckle

Let us denote by $I(\mathbf{r})$ the intensity of the wave measured at point $\mathbf{r}$ in a speckle pattern. In the statistical approach, 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

$$E(\mathbf{r},t) = \mathrm{Re}[E(\mathbf{r})\,\exp(-i\omega t)] \tag{6.1}$$

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. A scattering sequence is defined by a number of scattering events and the position of each scattering events. In this picture, the complex amplitude of the field can be written

$$E(\mathbf{r}) = \sum_{\mathcal{S}} A_{\mathcal{S}}(\mathbf{r})\,\exp[i\phi_{\mathcal{S}}(\mathbf{r})] \tag{6.2}$$

where $\mathcal{S}$ denotes any scattering sequence that starts on the entry surface of the medium and ends up at the observation point $\mathbf{r}$. This expansion of the field in terms of scattering sequences has been justified in chapter 3. 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. (6.2) are usually denoted by *random phasor sums* [18, 19].

The statistical properties of the intensity produced by a field of the form (6.2) 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 (complex) Gaussian statistics.

6.2 Amplitude distribution function

The sum (6.2) 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:

$$\begin{aligned} \langle X^2\rangle &= \sum_{\mathcal{S}}\sum_{\mathcal{S}'} \langle A_{\mathcal{S}}(\mathbf{r}) A_{\mathcal{S}'}(\mathbf{r})\rangle \langle \cos\phi_{\mathcal{S}}(\mathbf{r}) \cos\phi_{\mathcal{S}'}(\mathbf{r})\rangle \\ &= \sum_{\mathcal{S}} \langle A^2_{\mathcal{S}}(\mathbf{r})\rangle \langle \cos^2\phi_{\mathcal{S}}(\mathbf{r})\rangle \\ &= \frac{1}{2}\sum_{\mathcal{S}} \langle A^2_{\mathcal{S}}(\mathbf{r})\rangle \end{aligned} \tag{6.3}$$

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 (6.2), 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:

$$p(X,Y) = \frac{1}{2\pi\sigma^2} \exp\left(-\frac{X^2+Y^2}{2\sigma^2}\right) \,. \tag{6.4}$$

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

$$\begin{aligned} p(A) &= \frac{A}{\sigma^2} \exp\left(-\frac{A^2}{2\sigma^2}\right) \quad \text{for} \quad A > 0 \\ p(A) &= 0 \quad \text{for} \quad A < 0 \,. \end{aligned} \tag{6.5}$$

6.3 Intensity distribution function

The probability density of the intensity $I(\mathbf{r}) = |E(\mathbf{r})|^2$ directly follows from Eq. (6.5), using the relationship

$$p(I) = p(A = \sqrt{I}) \frac{dA}{dI} = \frac{1}{2\sqrt{I}} p(A = \sqrt{I}) \; . \tag{6.6}$$

We finally obtain:

$$\begin{aligned} p(I) &= \frac{1}{\langle I \rangle} \exp\left(-\frac{I}{\langle I \rangle}\right) \quad \text{for} \quad I > 0 \\ p(I) &= 0 \quad \text{for} \quad I < 0 \; . \end{aligned} \tag{6.7}$$

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 $I = 0$.

6.4 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:

$$\langle I^2 \rangle = \int_0^\infty I^2 \, p(I) \, dI = 2 \langle I \rangle^2 \; . \tag{6.8}$$

We end up with

$$\mathrm{Var}(I) = \langle I \rangle^2 \tag{6.9}$$

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

$$\frac{\sigma_I}{\langle I \rangle} = 1 \; . \tag{6.10}$$

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.

6.5 Intensity statistics of unpolarized light

Expression (6.7) 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. (6.7). Noticing that $\langle I_\alpha \rangle = \langle I_\beta \rangle = \langle I \rangle / 2$, the convolution product takes the form:

$$p(I) = \left(\frac{2}{\langle I \rangle}\right)^2 \int_0^I \exp\left[-\frac{2(I-x)}{\langle I \rangle}\right] \exp\left(-\frac{2x}{\langle I \rangle}\right) dx \tag{6.11}$$

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

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

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) [20].

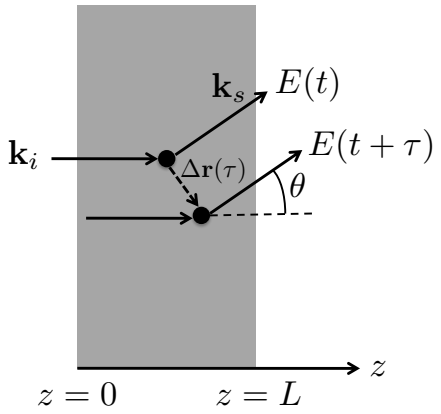

Figure 7.1: Dynamic light scattering in the single scattering regime. $\Delta\mathbf{r}(\tau)$ is the displacement of a scatterer between times t and $t+\tau$. This displacement induces a phase shift between the scattered fields $E(t)$ and $E(t+\tau)$.

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

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

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

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

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

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

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 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 $G_1(\tau)$. When the fields scattered by different particles can be considered as uncorrelated, a simple relation exist between $G_2(\tau)$ and $G_1(\tau)$.

Using Eq. (7.1), the intensity correlation function can be written

$$G_2(\tau) = |S(q)|^4\, \frac{|E_0|^4}{r^4} \sum_{j,k,l,m} \langle \exp\left[-i\mathbf{q}\cdot\mathbf{r}_j(t)\right] \exp\left[i\mathbf{q}\cdot\mathbf{r}_k(t)\right] \exp\left[-i\mathbf{q}\cdot\mathbf{r}_l(\tau)\right] \exp\left[i\mathbf{q}\cdot\mathbf{r}_m(\tau)\right]\rangle\ . \tag{7.8}$$

Assuming that the motions of different particles are uncorrelated, the only non vanishing terms in the average are those corresponding to $j = k = l = m$, to $j = k$ and $l = m$ with $k \neq l$, and $j = m$ and $k = l$ with $j \neq k$, so that

$$G_2(\tau) = |S(q)|^4 \frac{|E_0|^4}{r^4} \left[N^2 + \sum_{j\neq k} \left\langle \exp\left[-i\mathbf{q}\cdot\Delta\mathbf{r}_j(\tau)\right]\right\rangle \left\langle \exp\left[i\mathbf{q}\cdot\Delta\mathbf{r}_k(\tau)\right]\right\rangle\right]. \tag{7.9}$$

Since the average is the same for all particles, we obtain

$$G_2(\tau) = |S(q)|^4 \frac{|E_0|^4}{r^4} \left[N^2 + N(N-1) \left\langle \exp\left[-i\mathbf{q} \cdot \Delta\mathbf{r}(\tau)\right] \right\rangle^2 \right]. \tag{7.10}$$

Using Eq. (7.3), and assuming a large number of particles $N \gg 1$, the above expression can be cast in the following form:

$$G_2(\tau) = |G_1(0)|^2 + |G_1(\tau)|^2. \tag{7.11}$$

This relation, known as the Siegert relation, shows that the intensity correlation function can be obtained from the square modulus of the field correlation function. In terms of normalized correlation functions $g_1(\tau) = \langle E(t)\, E^*(t+\tau)\rangle / \langle |E(t)|^2 \rangle$ and $g_2(\tau) = \langle I(t)\, I(t+\tau)\rangle / \langle I(t)\rangle^2$, the Siegert relation simplifies into

$$g_2(\tau) = 1 + |g_1(\tau)|^2. \tag{7.12}$$

This relationship is frequently used in the analysis of DLS experiments. It has been derived here in the single scattering regime. In the multiple scattering regime, this relations holds provided that the field can be considered as a Gaussian variable (see chapter 6). In this case the intensity correlation function factorizes into products of second-order correlation functions, this factorization leading to the Siegert relation. Note that taken at $\tau = 0$, the Siegert relation (7.11) leads to $\langle I^2 \rangle = 2\langle I \rangle^2$, which is a feature of the Rayleigh statistics derived in chapter 6.

7.3 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

$$E(t) = E_0 \sum_{n=1}^{\infty} \sum_{\mathcal{S}_n} A_{\mathcal{S}_n}(t)\, \exp[i\phi_{\mathcal{S}_n}(t)] \tag{7.13}$$

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:

$$G_1(\tau) = |E_0|^2 \sum_{n} \sum_{\mathcal{S}_n} \sum_{n'} \sum_{\mathcal{S}'_{n'}} \langle\, A_{\mathcal{S}_n}(t) A_{\mathcal{S}'_{n'}}(t+\tau)\, \exp[i\phi_{\mathcal{S}_n}(t)] \exp[-i\phi_{\mathcal{S}'_{n'}}(t+\tau)] \,\rangle \tag{7.14}$$

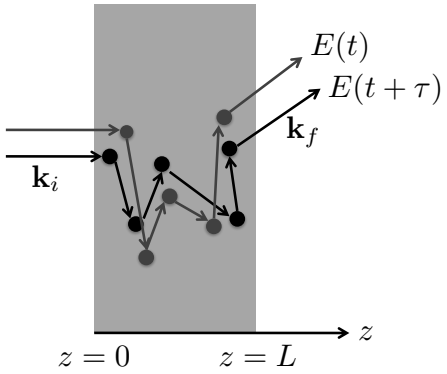

Figure 7.2: Schematic representation of the field resulting from a scattering sequence with multiple scattering events, at two differnet times t and $t+\tau$. As a consequence of the motion of scatterers, the accumulated phase shifts along the sequences at t and $t+\tau$ are different,

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

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle\, A_{\mathcal{S}_n}(t) A_{\mathcal{S}_n}(t+\tau) \,\rangle \, \langle \exp[i\phi_{\mathcal{S}_n}(t) - i\phi_{\mathcal{S}_n}(t+\tau)] \,\rangle. \tag{7.15}$$

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

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \, \langle \exp[i\phi_{\mathcal{S}_n}(t) - i\phi_{\mathcal{S}_n}(t+\tau)] \,\rangle \,. \tag{7.16}$$

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

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \left\langle \exp\left[i \sum_{j=1}^{n} \mathbf{q}_j \cdot \Delta \mathbf{r}_j(\tau) \right] \right\rangle \,. \tag{7.17}$$

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

$$G_1(\tau) = |E_0|^2 \sum_n \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2 \right) \right\rangle \tag{7.18}$$

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

$$P(n) = \sum_{\mathcal{S}_n} \langle A^2_{\mathcal{S}_n} \rangle \tag{7.19}$$

as the fraction of the incident power that has undergone n scattering events, and rewrite Eq. (7.18) in the form

$$G_1(\tau) = |E_0|^2 \sum_n P(n) \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2 \right) \right\rangle . \tag{7.20}$$

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

$$\begin{aligned} \left\langle \exp\left(-D_B\,\tau \sum_{j=1}^{n} q_j^2 \right) \right\rangle &\simeq \left\langle 1 - D_B\,\tau \sum_{j=1}^{n} q_j^2 \right\rangle \\ &= 1 - D_B\,\tau \sum_{j=1}^{n} \langle q_j^2 \rangle \\ &= 1 - n\,D_B\,\tau \langle q_j^2 \rangle \\ &\simeq \exp(-n\,D_B\,\tau \langle q_j^2 \rangle) . \end{aligned} \tag{7.21}$$

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

$$\langle q_j^2 \rangle = 2k_0^2 \langle 1 - \cos\theta \rangle = 2k_0^2 (1 - g) = 2k_0^2 \frac{\ell_s}{\ell_t} \tag{7.22}$$

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

$$G_1(\tau) = |E_0|^2 \sum_n P(n) \exp\left(-2k_0^2 \frac{\ell_s}{\ell_t} n\,D_B\,\tau \right). \tag{7.23}$$

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:

$$g_1(\tau) = \int_0^\infty P(s) \exp\left(-2\frac{\tau}{\tau_0}\frac{s}{\ell_t}\right) ds \tag{7.24}$$

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) [21].

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. (7.24) is mathematically a Laplace transform, on gets $g_1(\tau)$ directly from the solution of the RTE or diffusion equation in the Laplace domain [21].

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. (7.24) can be performed analytically (not shown) [21]. 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

$$g_1(\tau) \simeq \exp(-\gamma x) \tag{7.25}$$

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.

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

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

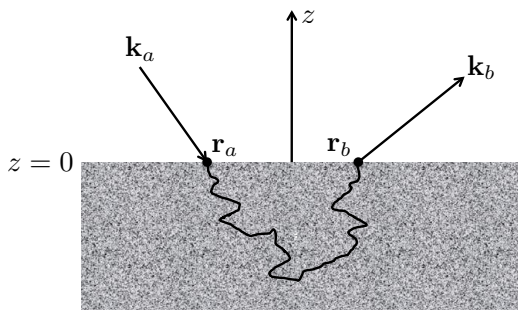

Figure 8.1: Reflection on a scattering medium illuminated by an incident plane wave with wavevector $\mathbf{k}_a$. One observes the far-field intensity reflected along direction $\mathbf{k}_b$.

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 [22]). 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, that is qualitatively introduced in chapter 3, is expected to give the leading contribution, corresponding to the diagrammatic representation in Fig. 8.2.

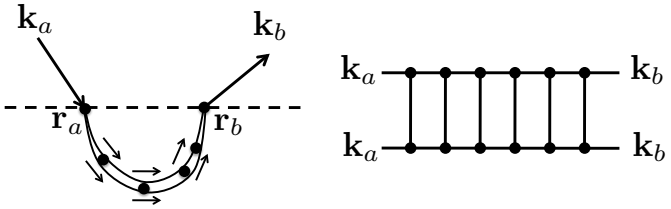

Figure 8.2: Graphical (left) and diagrammatic (right) representations of the diffuse reflection process in the ladder approximation.

In the ladder approximation, we can write the intensity correlator of the amplitude propagator as (see Eq. 3.33)

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

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. [22])

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

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

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

Making use again of (8.6) to simplify the correlator of the amplitude propagator leads to

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

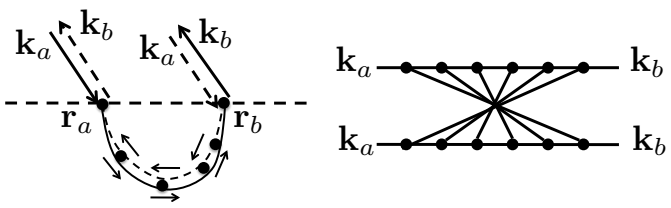

Figure 8.3: Graphical (left) and diagrammatic (right) representations of the reciprocal scattering sequences contributing to coherent backscattering. The diagram on the right is the maximally-crossed diagram.

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:

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

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 calculate explicitly 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

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

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

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

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 Ref. [11] 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

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

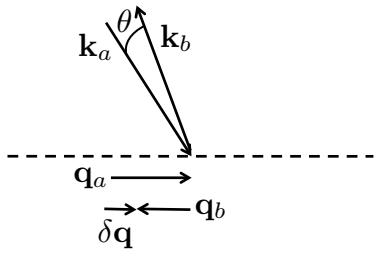

Figure 8.4: Wavevectors involved in the computation of the backscattered diffuse intensity.

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.

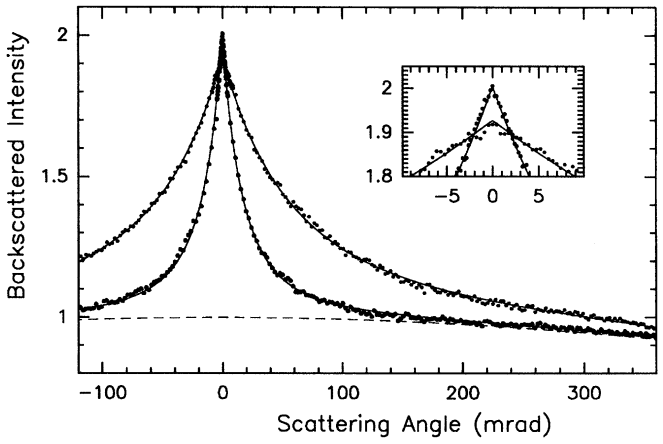

Figure 8.5: Experimental evidence of coherent backscattering of light from a strongly scattering medium. Narrow cone: $k\ell_t \simeq 23$. Wide cone: $k\ell_t \simeq 6$ (with k the wavenumber in the medium). From D. Wiersma *et al.*, Phys. Rev. Lett. **74**, 4193 (1995).

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.

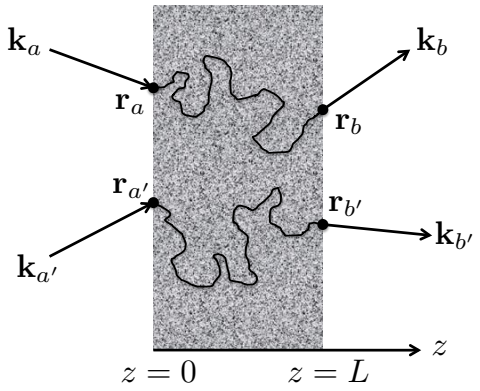

Figure 9.1: Geometry used for the calculation of the angular correlation function of the intensity transmitted through a slab of scattering material.

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 (or equivalently that transport can be described in the ladder approximation introduced qualitatively in chapter 3). 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. [22])

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

[1] A feature of Gaussian variables is that high-order correlation functions can always be factorized into products of second-order correlation function. An example is the Siegert relation derived in chapter 7.

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 $z = L$:

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

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. 3.33):

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

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 C. 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 C). 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

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

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

Appendices

Appendix A

Scattering matrix for polarized light

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

A.1 Scattering matrix

Following the traditon to work with the electric fied, the complex amplitude of the incident plane wave is of the form

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

with $\mathbf{E}_0 = E_0 \, \mathbf{e}_0$, the unit vector $\mathbf{e}_0$ describing the direction of polarization. The far field amplitude of the scattered field can be written

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

which defines the scattering matrix $\mathbf{S}(\mathbf{u})$. 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:

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

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

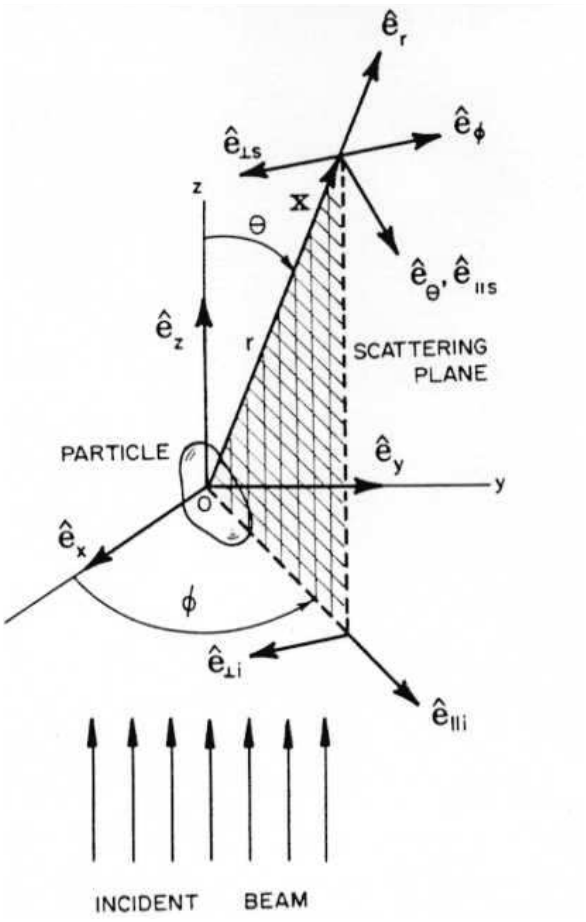

Figure A.1: Geometry used to define the scattering matrix for polarized light. From Ref. [3].

Using these bases, the scattering matrix $\mathbf{S}(\mathbf{u})$ is usually written in the form [3]

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

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:

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

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.

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

A.3 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 [3]:

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

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].

Appendix B

Examples of phase functions

We assume that the phase function depends only on $\cos\Theta = \mathbf{u} \cdot \mathbf{u}'$.

B.1 Isotropic scattering

For isotropic scattering, the simplest model, the phase function is a constant: $p(\cos\Theta) = 1$. This gives an anisotropy factor $g = 0$.

B.2 Rayleigh scattering

For particles much smaller than the wavelength, the electric dipole approximation applies. This is the regime of Rayleigh scattering. For an unpolarized incident field, the Rayleigh phase function is

$$p(\cos\Theta) = \frac{3}{4}(1 + \cos^2\Theta) \; . \tag{B.1}$$

This phase function also leads to $g = 0$.

B.3 Henyey-Greenstein model

The Henyey-Greenstein phase function is parametrized only by the anisotropy factor g. Its expression is

$$p(\cos\Theta) = \frac{1 - g^2}{\sqrt{(1 + g^2 - 2\, g\, \cos\Theta)^3}} \; . \tag{B.2}$$

The Henyey-Greenstein phase function is a convenient model to handle light propagation in biological tissues.

B.4 Mie scattering

The Mie theory provides an exact solution to the problem of scattering of an incident plane wave by a homogeneous spherical particle. From the knowledge of the refractive index of the particle and of the host medium, of the wavelength in the host medium, and of the particle radius, the Mie theory gives explicit expresssions of the cross sections and the phase function of a single particle in the form of series. For an ensemble of identical particles in the independent scattering regime, the phase function is that of a single particle. For a mixture of different particles, one needs to average the phase function according to the relative number density of each type of particle.

User-friendly solvers for the Mie theory are easily found on the internet.

B.5 Expansion on Legendre polynomials

An arbitrary phase function $p(\cos\Theta)$ can be expanded on the basis of Legendre polynomials $P_n(x)$, in the form

$$p(\cos\Theta) = \sum_{n=0}^{\infty} a_n \, P_n(\cos\Theta) \tag{B.3}$$

where the a_n are coefficients that are specific for each phase function. In practice, the series has to be truncated, and the number of terms to get convergence depends on the degree of anisotropy of the phase function.

Using the normalization of the phase function, and the definition of the anisotropy parameter g, we can show that $a_0 = 1$ and $a_1 = 3g$. For the particular case of the Henyey-Greenstein phase function, we can calculate the a_n explicitly, and we have $a_n = (2n+1)\, g^n$.

Appendix C

Diffusion equation and boundary conditions at an interface

In this appendix 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). We assume that the interface is illuminated by a plane wave at normal incidence (see Fig. C.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 $\ell_s = 1/\mu_s$.

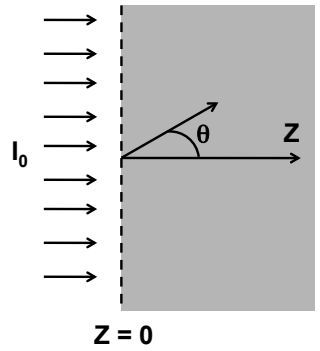

Figure C.1: Geometry used to study the boundary conditions in the diffusion approximation. The medium $z < 0$ is non scattering, while the medium $z > 0$ is assumed to be strongly scattering. The interface $z = 0$ is illuminated by a plane wave with intensity I_0 (W.m^{-2}).

C.1 Diffusion equation with collimated illumination

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. C.1 in which the specific intensity only depends on z and $\mathbf{u}$ due to translationnal invariance along the $x-y$ directions, we have

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

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. (C.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:

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

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

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

C.2 Boundary condition at $z=0$

In order to get a boundary condition in terms of the energy density, we can use the fact that the diffuse incoming flux vanishes at the interface (we follow the procedure in Ref. [16]). In terms of the diffuse specific intensity, this reads as

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

where the angular integration is over directions satisfying $\mathbf{u}\cdot\mathbf{e}_z>0$. Under the P_1 approximation, we have (Eq. 5.9):

$$I_{\rm d}(z,\mathbf{u})=\frac{v_E\,U(z)}{4\pi}+\frac{3}{4\pi}\,\mathbf{q}(z)\cdot\mathbf{u}\;. \tag{C.5}$$

Inserting this expansion into Eq. (C.4) leads to

$$\frac{v_E\, U(z=0)}{4\pi} \int_{2\pi} \mathbf{u}\cdot\mathbf{e}_z\, d\Omega + \frac{3}{4\pi}\int_{2\pi} \mathbf{q}(z=0)\cdot\mathbf{u}\,(\mathbf{u}\cdot\mathbf{e}_z) d\Omega = 0\ . \tag{C.6}$$

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

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

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

$$U(z=0) - \frac{2}{3}\frac{1}{\mu_a+\mu_s'}\,\frac{dU}{dz}(z=0) + \frac{2\mu_s\, g}{v_E(\mu_a+\mu_s')}\, I_{\rm b}(z=0) = 0\ . \tag{C.7}$$

In the absence of the collimated source term $I_{\rm b}(z=0) = 0$, the boundary condition simpifies into

$$U(z=0) - \frac{2}{3}\frac{1}{\mu_a+\mu_s'}\,\frac{dU}{dz}(z=0) = 0\ . \tag{C.8}$$

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 $z_0 = (2/3)(\mu_a+\mu_s')^{-1}$, known as the extrapolation distance. When absorption can be neglected we simply have $z_0 = (2/3)\,\ell_t$. Imposing $U(z) = 0$ for $z = -z_0$ is equivalent to imposing (C.8).

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. [23, 24, 12] and [1] (chap. 25).

Appendix D

Diffuse transmission through a slab

In this appendix 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.1). We assume a non absorbing medium ($\mu_a = 0$).

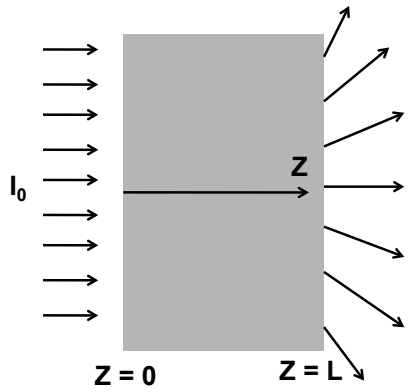

Figure D.1: Diffuse transmission through a slab with thickness L. The interface $z = 0$ is illuminated by a plane wave with intensity I_0 (W.m^{-2}).

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

$$\frac{d^2 U}{dz^2} = -\frac{3\mu_s^2}{v_E} I_{\mathrm{b}}(z) \tag{D.1}$$

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

The general solution is the sum of the solution to $d^2 U/dz^2 = 0$ and a particular solution of

Eq. (D.1). Using $U_p(z) = (-3I_0/v_E)\exp(-\mu_s z)$ as a particular solution, we have

$$U(z) = C_1 z + C_2 - \frac{3I_0}{v_E}\,\exp(-\mu_s z) \tag{D.2}$$

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. C.7)

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

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

$$U(z=L) + \frac{2}{3}\frac{1}{\mu_a + \mu_s'}\,\frac{dU}{dz}(z=L) - \frac{2\mu_s\, g}{v_E(\mu_a + \mu_s')}\,I_{\rm b}(z=L) = 0\ . \tag{D.4}$$

Inserting Eq. (D.2) 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

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

where $z_0 = (2/3)\,\ell_t$.

The transmitted flux (unit W.m^{-2}) is

$$\phi = -D\,\frac{dU}{dz}(z=L) \tag{D.6}$$

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.5) and (D.6):

$$T = \frac{5}{3}\frac{\ell_t}{L + 2z_0} \simeq \frac{5}{3}\frac{\ell_t}{L} \tag{D.7}$$

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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