

Light Propagation in Chiral and Magnetochiral Random Media

by

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Abstract

Thesis Supervisor: Bart van Tiggelen
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Introduction and general presentation

The complex nature of wave transport can have several origins. These origins can generally be identified as some complex medium that supports wave propagation with some complex constraint. As examples of complex media that have recently attracted attention, one can mention cold atomic gases [1], strongly disordered media where Anderson localization can occur [2], photonic band gap materials [3] and natural environments, such as the Earth's crust [4] and biological tissues [5], intrinsically complex media that are now being studied using multiple scattering techniques. Much of this progress is supported by interdisciplinary collaborations, motivated mainly by several applications (for a recent review, see Ref. [6]). Another important source of complexity is broken symmetry. It imposes an important constraint to wave propagation in random media since it can dramatically influence observables of wave transport, giving rise to a new class of ondulatory phenomena. In this thesis we will focus on two different aspects of complexity in wave transport in random media: broken symmetries and Anderson localization. For this reason this thesis is divided in two parts.

In the first part, we theoretically investigate light propagation in inhomogeneous media exhibiting broken time-reversal and/or mirror symmetry. Media that lack mirror symmetry are called *chiral* media and for this reason they exhibit natural optical activity. The breaking of time-reversal symmetry in optical media, induced by an external magnetic field, is responsible for many magneto-optical effects, such as the Faraday effect. When mirror and time-reversal symmetries are simultaneously broken, a cross-effect between magnetic and natural optical activity, the so-called *magnetochiral effect*, can occur. The magnetochiral effect has recently

been observed, both in emission [7] and in refraction [8][9], and constitutes a fundamental issue for the understanding the symmetry principles that govern light propagation. In this thesis we will study light propagation in magnetochiral and chiral *inhomogeneous scattering media* by means of both numerical simulations and rigorous transport theory.

The second part is mainly dedicated to *Anderson localization in open systems*. The phenomenon of Anderson localization was originally conceived by P. W. Anderson as an “absence of diffusion” in wave transport due to interference effects in *infinite* media [10]. However, actual experiments are performed in *finite* media. In addition, experiments on wave transport are usually conducted in open media, i.e., media where waves can “leak” through the sample boundaries. As a result, the “eigenstates” of an open system are not bound states as in closed systems, but *resonances* with a finite energy width Γ or, equivalently, with a finite lifetime $t \sim 1/\Gamma$. In view of these facts, it is desirable to investigate how Anderson localization manifests itself in leakage-related quantities, such as resonance widths or, more interestingly, the statistical distribution of resonance widths $P(\Gamma)$ for many different configurations of the disorder. This is the main objective of the work presented in the second part of the thesis. We will also investigate how an external magnetic field affects $P(\Gamma)$ and suggest possible applications to random lasers.

Introduction et présentation générale

Le caractère complexe du transport ondulatoire peut avoir plusieurs origines. Ces origines sont en général dues à la complexité du milieu ou à quelques contraintes complexes à la propagation des ondes. Comme exemples de milieux complexes qui ont attirés récemment beaucoup d'attention, nous pouvons mentionner les nuages d'atomes froids [1], les milieux fortement désordonnés où la localisation d'Anderson peut se produire [2], les matériaux photoniques à bande interdite [3] mais aussi les milieux naturels, comme par exemple la croûte terrestre [4] et les tissus biologiques [5], qui sont des milieux intrinsèquement complexes et qui sont maintenant étudiés via les méthodes de la diffusion multiple. Le progrès en ce domaine est plutôt dû aux collaborations multidisciplinaires motivées par de nombreuses applications (pour une oeuvre de révision récente, voir Ref. [6]). La symétrie brisée est aussi une source importante de complexité. Elle impose des contraintes importantes à la propagation des ondes dans les milieux aléatoires car elle peut influencer dramatiquement les observables du transport, déterminant de nouveaux phénomènes ondulatoires. Dans cette thèse nous nous intéressons à deux différents aspects de la propagation des ondes dans les milieux désordonnés : les symétries brisées et la localisation d'Anderson. Pour cette raison, la thèse est divisée en deux parties.

Dans la première partie, nous ferons une étude de la propagation de la lumière dans les milieux inhomogènes qui brisent la symétrie du renversement du temps et/ou miroir. Les milieux où la symétrie miroir est absente sont appelés milieux *chiraux* et pour cette raison ils présentent l'activité optique naturelle. La brisure de symétrie par renversement du temps dans les milieux optiques, induite par la présence d'un champ magnétique externe, est responsable de plusieurs

effets magnéto-optiques, comme l'effet Faraday. Quand les symétries miroir et par renversement du temps sont simultanément brisées, un effet croisé entre l'activité optique naturelle et l'activité optique magnétiquement induite, l'effet *magnéto-chiral*, peut se produire. L'effet magnéto-chiral a été récemment observé en émission [7] et en réfraction [8][9], et constitue un aspect important pour la compréhension des principes de symétrie qui gouvernent la propagation de la lumière. Dans cette thèse nous étudierons la propagation de la lumière dans les milieux *inhomogènes* magnéto-chiraux et chiraux en *diffusion*, en utilisant des simulations numériques et la théorie du transport.

La deuxième partie de la thèse est plutôt consacrée à la *localisation d'Anderson dans les milieux ouverts*. Le phénomène de la localisation d'Anderson a été conçu par P. W. Anderson à l'origine comme « l'absence de diffusion » en transport ondulatoire en raison des effets d'interférence dans les milieux aléatoires *infinis* [10]. Cependant, les expériences réelles sont faites dans les milieux *finis*. En outre, les expériences de transport ondulatoire sont fréquemment réalisées dans les milieux ouverts, i.e. les milieux où les ondes peuvent « fuir » à travers les bords de l'échantillon. Comme conséquence, les « états propres » d'un système ouvert ne sont pas états liés comme dans les milieux fermés, mais *résonances* avec une largeur finie en énergie Γ ou, équivalamment, avec un temps de vie fini $t \sim 1/\Gamma$. Dans ce contexte, il est souhaitable d'étudier comment la localisation d'Anderson se manifeste dans les quantités liées à la fuite, comme les largeurs de résonance ou, plus intéressant encore, dans la statistique des largeurs de résonance $P(\Gamma)$ pour plusieurs réalisations du désordre. Ceci est le principal objectif du travail présenté dans la deuxième partie de la thèse. Nous nous intéressons également à l'étude de la façon dont la présence d'un champ magnétique externe affecte $P(\Gamma)$ et nous proposons des applications possibles dans le domaine des lasers aléatoires.

Part I

Light propagation in chiral and magneto-chiral random media

Plan of part I

The first part of the thesis is devoted to the study of light propagation in inhomogeneous media exhibiting two broken symmetries, namely the time-reversal symmetry and mirror symmetry. It is organized as follows. Chapter 1 is devoted to an overview of light propagation in homogeneous media exhibiting broken symmetries, both mirror and time-reversal symmetries. In Chapter 2, we will consider light scattering by single point-dipoles, i.e. particles much smaller than the optical wavelength, an approximation which we will employ throughout this thesis to model multiple wave scattering. Chapter 3 contains a brief discussion of some aspects of rigorous transport theory to be applied in the thesis. In Chapter 4, we will introduce a model to describe light propagation in inhomogeneous magnetochiral media and suggest that the magnetochiral effect can be used to define optical parameters to “measure” the degree of chirality of scattering objects. Finally, in Chapter 5 we will apply rigorous transport theory to describe light propagation in chiral and magnetochiral random media focusing on the impact of broken symmetries.

Plan de la partie I

La première partie de la thèse est consacrée à l'étude de la propagation de la lumière dans les milieux inhomogènes où les symétries par renversement du temps et/ou miroir sont brisées. Elle est organisée de la façon suivante. Le Chapitre 1 est dédié à la propagation de la lumière dans les milieux homogènes avec les symétries brisées (symétries miroir et renversement du temps). Au Chapitre 2 nous considérons la diffusion de la lumière par des dipôles ponctuels, i.e. pour les particules beaucoup plus petites que la longueur d'onde optique, une approximation que nous utiliserons tout au long de la thèse. Le Chapitre 3 contient une brève discussion sur quelques aspects de la théorie rigoureuse du transport, que nous allons également utiliser dans la thèse. Au chapitre 4 nous introduisons un modèle pour décrire la propagation de la lumière dans les milieux magnéto-chiraux inhomogènes. Nous allons suggérer aussi que l'effet magnéto-chiral peut être utilisé pour définir de nouveaux paramètres optiques pour « mesurer » le degré de chiralité associé aux objets diffuseurs. Finalement, au Chapitre 5 nous allons employer les concepts de la théorie rigoureuse du transport pour décrire la propagation de la lumière dans les milieux aléatoires chiraux et magnéto-chiraux, en nous focalisant sur l'impact des symétries brisées.

Chapter 1

Light propagation in chiral and/or magneto-optical homogeneous media

1.1 Propagation in magneto-optical media

1.1.1 The role of symmetry

The basis for understanding the propagation of classical electromagnetic waves is provided by the Maxwell equations. They are believed to provide a complete description of the spatial and temporal evolution of the macroscopic electromagnetic fields $\mathbf{E}$ and $\mathbf{B}$. The propagation of electromagnetic radiation affects dramatically the distribution of charges and currents inside matter which, at a microscopic level, can be described by the polarization density vector $\mathbf{P}$ and the magnetization $\mathbf{M}$. In a simple classical model, bound electrons in matter move under the influence of an electromagnetic wave, resulting in a polarization of the atom described by $\mathbf{P}$. These moving electrons, under influence of the magnetic field $\mathbf{B}$ of the electromagnetic wave, are also deflected by the Lorentz force, which alters the polarization $\mathbf{P}$. In addition, the magnetic field $\mathbf{B}$ induces the formation of microscopic magnetic moments which result in a macroscopic magnetization $\mathbf{M}$ ¹.

In a (dispersive) dielectric medium with magnetic permeability $\mu = 1$ (relative to that of

¹This effect is, however, negligible in the optical region.

the vacuum μ_0) and the relative dielectric constant $\varepsilon(\mathbf{r}, \omega)$, the Maxwell equations are given by:

$$\nabla \times \mathbf{E} = -\partial \mathbf{B} / \partial t \quad (1.1)$$

$$\nabla \times \mathbf{B} = \mu_0 \partial \mathbf{D} / \partial t \quad (1.2)$$

$$\nabla \cdot \mathbf{E} = 0 \quad (1.3)$$

$$\nabla \cdot \mathbf{B} = 0, \quad (1.4)$$

where $\mathbf{D} = \varepsilon \mathbf{E}$ is the electric displacement. By the elimination of $\mathbf{B}$, one obtains the Helmholtz equation containing only the electric field $\mathbf{E}$:

$$-\frac{\varepsilon(\mathbf{r}, \omega) \omega^2}{c_0^2} \mathbf{E}(\mathbf{r}, \omega) + \nabla \times \nabla \times \mathbf{E}(\mathbf{r}, \omega) = 0, \quad (1.5)$$

where c_0 is the velocity of light in the vacuum. The complete solution of Maxwell's equations is only possible if the link between the macroscopic electromagnetic fields $\mathbf{E}$ and $\mathbf{B}$ and the microscopic parameters $\mathbf{P}$ and $\mathbf{M}$ is known. The so-called constitutive relations represent such a link. The form of these constitutive relations is subject to major constraints due to the symmetry relations: parity and time-reversal. Indeed, these relations must take into account the fact that the electric field $\mathbf{E}$ and the polarization $\mathbf{P}$ change sign upon space inversion (they are called polar or parity-odd vectors) and the magnetic field $\mathbf{B}$ and the magnetization $\mathbf{M}$ do not (they are called axial vectors or pseudovectors). In addition, they must be consistent with the fact that both $\mathbf{B}$ and $\mathbf{M}$ change sign under time-reversal whereas $\mathbf{E}$ and $\mathbf{P}$ do not. One of the symmetry-allowed forms for the constitutive relations is [11]:

$$\mathbf{P} = \chi_0 \mathbf{E} + \chi_1 \frac{\partial \mathbf{E}}{\partial t} \times \mathbf{B} + \chi_2 (\mathbf{B} \cdot \mathbf{B}) \frac{\partial^2 \mathbf{E}}{\partial t^2} + \chi_3 \left(\frac{\partial^2 \mathbf{E}}{\partial t^2} \cdot \mathbf{B} \right) \mathbf{B} + \dots \quad (1.6)$$

For the sake of simplicity, we assume that the medium is isotropic and consequently all the parameters χ_n are scalars and not tensors. In the presence of a low frequency, external magnetic field $\mathbf{B}_{ext}$ with magnitude much larger than the electromagnetic field $\mathbf{B}$ itself, the constitutive relation (1.6) assumes the simple linearized form:

$$\mathbf{P}(\omega) = \chi(\omega, \mathbf{B}_{ext}) \cdot \mathbf{E}(\omega), \quad (1.7)$$

with $\chi(\omega, \mathbf{B}_{ext})$ the electrical susceptibility tensor:

$$\chi_{ij}(\omega, \mathbf{B}_{ext}) = \chi_0 \delta_{ij} + \chi_1 \omega i \epsilon_{ijk} B_k - \chi_2 \omega^2 B^2 \delta_{ij} - \chi_3 \omega^2 B_i B_j, \quad (1.8)$$

where ϵ_{ijk} is the anti-symmetric Levi-Civita tensor. In anisotropic media, the dielectric constant is a second-rank tensor related to the susceptibility tensor according to:

$$\varepsilon_{ij} \equiv \delta_{ij} + \chi_{ij}. \quad (1.9)$$

The complex tensor describing the index of refraction is given by:

$$n_{ij} = \sqrt{\varepsilon_{ij}}. \quad (1.10)$$

At optical frequencies the influence of an external magnetic field in the dielectric tensor is very small and can thus be treated as a small perturbation. In addition, it is an excellent approximation to set the relative magnetic permeability μ equal to one in the same region of the spectrum provided the medium is achiral. Under these assumptions, we can expand the dielectric tensor (1.8) up to first order in $\mathbf{B}_{ext}$:

$$\varepsilon_{ij}(\mathbf{B}_{ext}) = \varepsilon_0 \delta_{ij} + \varepsilon_F \Phi_{ij}, \quad (1.11)$$

where $\varepsilon_0 \equiv n_0^2$ is the dielectric constant without magnetic field and the antisymmetric, Hermitian tensor Φ_{ij} is defined according to:

$$\Phi_{ij} = i \epsilon_{ijk} \widehat{B}_k. \quad (1.12)$$

For simplicity, in the following we will denote the external magnetic field by $\mathbf{B}$. The dimensionless parameter ε_F is given by:

$$\varepsilon_F(\omega) = 2n_0 V(\omega) c_0 B / \omega, \quad (1.13)$$

where $V(\omega)$ is the so-called Verdet constant, and which depends in general on frequency.

As is true in other branches of physics, symmetry laws govern electromagnetic propagation in matter subject to magnetic fields in a crucial way. This statement can be appreciated by studying the symmetry properties of the dielectric tensor (1.9). Indeed, symmetry imposes that the dielectric tensor, up to linear order in the magnetic field, must obey the following relations [12]:

$$\varepsilon_{ij}(\mathbf{B}) = \varepsilon_{ji}(-\mathbf{B}) \neq \varepsilon_{ji}(\mathbf{B}), \quad (1.14)$$

which are the optical equivalent of the Onsager relations in thermodynamics [13]. The first equality follows from the fact that the whole system “matter+external magnetic field” is invariant under time-reversal (recall that $\mathbf{B}$ changes its sign upon such an operation). The inequality implies that the magnetic field breaks time-reversal symmetry in the subsystem composed of matter only.

The aim of magneto-optics is to investigate the influence of an external magnetic field on electromagnetic propagation in matter, which can be described by an index of refraction that depends on the external magnetic field, as shown in Eqs. (1.8, 1.9 and 1.10). Different magneto-optical effects are described by the different χ_n coefficients in Eq. (1.8), as will be detailed in the following, showing that magneto-optics can be regarded as a particular case of nonlinear optics.

1.1.2 Magneto-optical effects in homogeneous media

The Faraday effect and the Verdet constant

The *Faraday effect* or more technically the magnetically-induced optical rotation is associated with the χ_1 term in Eq. (1.8), which is proportional to the Verdet constant, $\chi_1 = 2V/\omega^2$. The origin of the Faraday effect can be understood by considering the dispersion relation of photons propagating in a magneto-optical medium. This can be accomplished by inserting Eq. (1.7) into Eq. (1.8), resulting in:

$$\omega(k) = \frac{kc_0}{n_0} + \frac{\varepsilon_F c_0}{2n_0^3} \sigma(\hat{\mathbf{k}}) \hat{\mathbf{B}} \cdot \mathbf{k}, \quad (1.15)$$

where Eqs. (1.9), (1.11) and (1.13) have been used; $\mathbf{k}$ is the wavevector and $\sigma(\hat{\mathbf{k}})$ denotes the state of polarization of the incident light: +1 for right and -1 for left circularly polarized light (abbreviated RCP and LCP light in the following)². The dispersion law (1.15) implies that the constant frequency surface is shifted by an amount $\varepsilon_F c_0 / 2n_0^3$ from the origin along or opposite to the magnetic field, depending on the state of polarization of the incident light as illustrated in Fig. (1-2). This reveals that the Faraday effect can be regarded as a manifestation of the Zeeman effect: the magnetic field, removing the degeneracy of two states of circular polarization $\sigma = \pm 1$, is responsible for the two distinct light velocities associated with each state of circular polarization³. As a consequence, for incident linearly polarized light, which can always be decomposed into two circularly polarized components, the polarization vector of the emergent radiation will be rotated by an angle $\alpha = VBL$ along the direction of the magnetic field, with L the length of the optical path inside the medium. This effect was discovered in 1846 by Faraday. It is important to emphasize that the sense of the Faraday rotation changes if the direction of the magnetic field relative to the one of the light propagation changes. The angle of rotation α is multiplied by two if the light undergoes the same optical path twice (i.e., in a round trip in the medium).

If the χ_1 coefficient in Eq. (1.8) has a nonvanishing imaginary part, the imaginary part of the index of refraction will also be slightly different for the two states of circular polarization, which will thus be differently absorbed. As a result, an incident linearly polarized light beam becomes elliptical. This effect, called *magnetic circular dichroism* or Faraday ellipticity, constitutes an important tool in chemical physics since it allows a detailed study of the excited electronic states of metallic compounds [15].

The Verdet constant, which shows up as the essential coupling parameter between the light and the magnetic field, usually depends on both the optical wavelength λ and the temperature T . One can usually distinguish diamagnetic and paramagnetic contributions to the Verdet constant.

The diamagnetic part, which occurs in every material, has a slight influence on the Faraday

²We will adopt throughout this thesis the Born&Wolf convention for circular polarization: RCP light corresponds to a clockwise rotation of the electric field vector for an observer looking in the direction from which the light is propagating whereas for LCP light the opposite situation applies, i.e., the rotation is anticlockwise [14].

³This is argument just applies for the diamagnetic contribution to the Faraday effect, which occurs in most materials. The origin of the Faraday effect in paramagnetic materials is more complicated.

Figure 1-1:

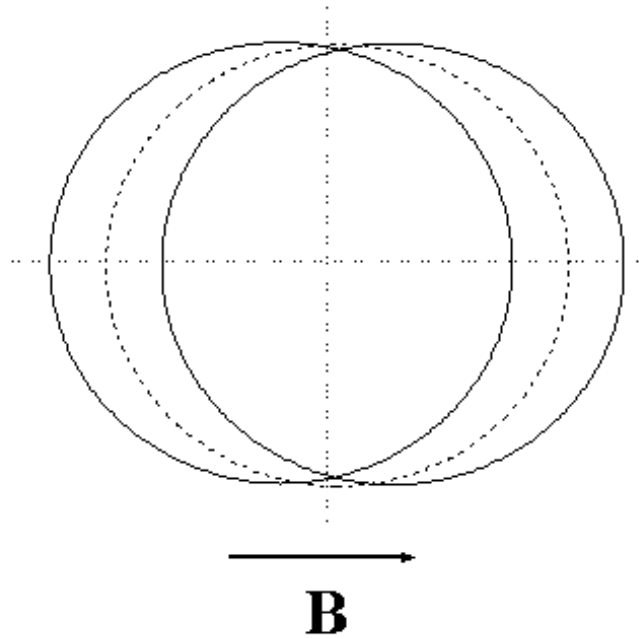


Figure 1-2: Dispersion law in a magneto-optical medium. The dotted circle is the constant frequency surface without magnetic field. In the presence of the magnetic field the degeneracy in the dispersion law is removed and its two solutions are associated with the two states of circular polarization, represented by full circles.

effect and typically gives a small, positive and temperature independent contribution to the Verdet constant [15]. The diamagnetic contribution to the Verdet constant finds its origin in the Zeeman effect, a fact that is evident in its wavelength dependence [15]:

$$V_{diamagnetic} \propto \lambda \left[\left(\frac{1}{\lambda^2 - \lambda_L^2} \right) - \left(\frac{1}{\lambda^2 - \lambda_R^2} \right) \right], \quad (1.16)$$

with λ the incident wavelength and λ_L and λ_R the wavelengths associated with left and right circularly polarized light.

On the other hand, the wavelength dependence of the Verdet constant for paramagnetic materials is more complicated than expressed in Eq. (1.16) since it is not directly related to the Zeeman effect as in the diamagnetic case [15]. The Verdet constant of paramagnetic materials typically exhibits a pronounced variation with temperature which is related to the temperature dependence of the paramagnetic susceptibility. Provided that the temperature is not too low ($T \lesssim 20K$), the paramagnetic contribution to the Verdet constant typically follows the Curie-Weiss $1/T$ temperature dependence of the paramagnetic susceptibility [16].

It is also possible to describe the Faraday effect classically, within the so-called “Becquerel model” [12]. Within this model, the interaction light-matter is treated classically, with the electron motion around the nucleus being described as a damped harmonic oscillator subject to the Lorentz force due the external magnetic field and to the electric force due the interaction with the radiation field.

It is interesting to mention that the Faraday effect was shown recently to be strongly enhanced inside the stop band of a photonic band-gap crystal embedded in Faraday active liquid [17].

The Cotton-Mouton effect

The terms associated with the coefficients χ_2 and χ_3 in Eq. (1.8) are quadratic in the magnetic field and, consequently, generate new magneto-optical effects though smaller than the Faraday effect. If the magnetic field is applied perpendicularly to the direction of propagation of the light beam, these terms induce a linear birefringence (the so-called *Cotton-Mouton* effect, discovered first in 1905). Physically, this effect finds its origin in a partial orientation of the molecules in a

magneto-optical medium. As a result, such a medium can be regarded as an uniaxial crystal with its optical axis parallel to the field. The coefficients χ_2 and χ_3 reflects this uniaxial symmetry and the relations (1.11) are no longer valid. It is worth mentioning that a phenomenologically identical effect to the Cotton-Mouton effect exists, namely the Kerr effect, but in this case the uniaxial symmetry is induced by an external electric field and not by a magnetic field.

Magnetoelectric Linear Birefringence, Magnetoelectric Jones Birefringence and Magnetoelectric Directional Anisotropy

Recently, Roth and Rikken have reported [18] the observation of a cross effect between the Cotton-Mouton and the Kerr effect. In this magneto-optical effect, called *Magnetoelectric linear birefringence* (MELB), a linear birefringence is induced by combining magnetic and electric fields. The MELB is an effect bilinear effect in $\mathbf{E}$ and $\mathbf{B}$, with $\mathbf{E}$ and $\mathbf{B}$ both perpendicular to the direction $\hat{\mathbf{k}}$ of propagation of light but not mutually parallel. It was first theoretically predicted by Pockels and later by Baranova [19], who argued that the MELB can only exist in media lacking mirror symmetry (so-called chiral media, see Sec. 1.1.2). However, a symmetry allowed expansion of the refractive index exists which generates a term linear both in $\mathbf{k}$, $\mathbf{E}$ and $\mathbf{B}$. This combination does not require the medium to be chiral and could thus occur in any material. Recent calculations predict that the MELB, as well as the Kerr and the Cotton-Mouton effect, can occur even in vacuum [20].

If $\mathbf{E}$ and $\mathbf{B}$ are both perpendicular to the direction of the incident light beam but now mutually parallel, another effect can occur: the *magnetoelectric Jones birefringence* (MEJB), which has also recently been observed [21]. As the MELB, the MEJB can also occur in any material [20]. Both MELB and MEJB result from a nonlocal optical response of the medium and have the same order of magnitude, as required by symmetry arguments [22], [23]. Their magnitudes are, however, smaller than those of the Kerr and Cotton-Mouton effect, which probably explains why they have only recently been observed.

In addition to MELB and MEJB, a third magnetoelectric optical effect was also recently observed: the *Magnetoelectric Directional Anisotropy* (MEA) [24]. The MEA manifests itself as a polarization-independent difference in the refractive index n for light propagating parallel or antiparallel to the vector $\mathbf{E} \times \mathbf{B}$, i.e., as a contribution to n of the form $\delta n \propto \hat{\mathbf{k}} \cdot (\mathbf{E} \times \mathbf{B})$.

The MEA has the same order of magnitude of MELB and can in fact be regarded as a relativistic correction to the Cotton-Mouton effect [24]. Moreover, recent calculations confirm the symmetry-based prediction that the MEA can occur in any medium, even in the quantum vacuum [25].

1.2 Light propagation in homogeneous chiral media

An object or medium is said to be chiral and to exhibit chirality if it does not possess mirror symmetry, i.e., those objects that cannot be superimposed on its mirror image by means of any solid rotation or translation. Chirality is an ubiquitous concepts in nature and plays a fundamental role in several areas of science, as will be discussed in more detail in Chapter 4.

In the present chapter, we will focus on only one of the manifestations of chirality in nature, though actually historically the first to be discovered: the *natural optical activity* (NOA). It is also important to emphasize that NOA constitutes, as the Faraday effect, an example of *gyrotropy*, i.e., the property that certain media can distinguish RCP and LCP light. NOA was first observed by Arago in 1811, who discovered that the plane of polarization of linearly polarized light propagating through a quartz crystal in the direction of the optical axis underwent a rotation proportional to the length of quartz it traversed. One year later, Biot observed that quartz can rotate the plane of polarization either for the right (dextro-rotatory power) or for the left (laevo-rotatory power), and that liquids, as well as quartz, can be optically active. However, Biot did not relate the sense of the optical rotation to the handedness of the quartz . Pasteur was the first to associate optical activity with chirality (which he called *dissymétrie*⁴) by observing that the optical rotation in solutions of sodium ammonium tartrate crystal has opposite sign depending on the handedness of the molecules in solution. Nevertheless, he erroneously associated the origin of this “dissymmetry” with the magnetic field, which he employed during the process of growing the crystals. In 1825, well before Maxwell’s electromagnetic theory of light was published, Fresnel interpreted circular birefringence as the difference in the velocity of propagation for RCP and LCP light, which explains the observed phenomenon of optical

⁴The french expression *dissymétrie* became generally supplanted by the term chirality, coined by Lord Kelvin in the beginning of the twentieth century: “I call any geometrical figure, or group of points, chiral, and say it has chirality if its image in a plane mirror, ideally realized, cannot be brought to coincide with itself. ”

rotation.

In order to describe the NOA in a quantitative way, let us consider light propagation in a homogeneous (i.e. the material parameters do not depend on the position $\mathbf{r}$), isotropic, nonmagnetic and gyrotropic medium. In the presence of gyrotropy (either natural or induced by an arbitrary external field $\mathbf{g}$), the constitutive relations can be written as follows [12]:

$$\mathbf{D} = \varepsilon_0[\mathbf{E} - if(\hat{\mathbf{k}} \times \mathbf{E}) - ig(\hat{\mathbf{g}} \times \mathbf{E})], \quad (1.17)$$

where the real-valued parameters f and g characterize the strengths of the natural and induced gyrotropic interactions, respectively. Since we are only interested in NOA, we put $\mathbf{g} = \mathbf{0}$ in the following. In tensorial form, Eq.(1.17) is written as:

$$\mathbf{D} = \tilde{\varepsilon} \cdot \mathbf{E}, \quad (1.18)$$

where the dielectric tensor:

$$\tilde{\varepsilon} = \varepsilon_0 \begin{pmatrix} 1 & if\gamma & -if\beta \\ -if\gamma & 1 & 0 \\ if\beta & 0 & 1 \end{pmatrix} \quad (1.19)$$

is Hermitian. The direction cosines of $\hat{\mathbf{k}}$ with respect to the y and z axes are designated by γ and β , respectively. We will not address here the issue of the origins of the form of Eq.(1.17) or (1.19), for which several microscopic models, both classical and quantum mechanical, can be found elsewhere [15]. We will only focus on the phenomenological manifestations in optics to which these relations give rise.

Assuming incident plane waves for the light field $\mathbf{E}$ in the medium

$$\mathbf{E}(\mathbf{r}, t) = E_0 \hat{\mathbf{e}} \exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)], \quad (1.20)$$

with propagation vector $\mathbf{k} = n(\omega/c)\hat{\mathbf{k}}$, $[\hat{\mathbf{k}} = (0, \beta, \gamma)]$, and substituting it into the Maxwell equations, one obtains a set of three linearly coupled equations for the components of the

electric field $\mathbf{E}$:

$$\begin{pmatrix} m^2 - 1 & -if\gamma & if\beta \\ if\gamma & m^2\gamma^2 - 1 & -m^2\gamma\beta \\ -if\beta & -m^2\gamma\beta & m^2\beta^2 - 1 \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = 0 \quad (1.21)$$

with $m^2 \equiv n^2/\varepsilon$. A nontrivial solution exists only if the determinant of the coefficient matrix vanishes. This yields a secular “Fresnel” equation quadratic in m^2 from which the indices of refraction $n^2(\hat{\mathbf{k}})$ in naturally optically active medium are obtained:

$$n_{\pm}^2 = \varepsilon(1 \pm f). \quad (1.22)$$

Rephrasing the above relation in terms of frequency, we obtain the following dispersion law in chiral media:

$$\omega(k) = \frac{kc_0}{n_0} \left[1 + f\sigma(\hat{\mathbf{k}}) \right]. \quad (1.23)$$

This dispersion relation is depicted in Fig. 1-3, which shows its isotropic character. In refraction, the effect expressed by Eq. (1.22) is also called natural birefringence, in absorption it is called natural dichroism.

Comparing Eq.(1.23) for a naturally optically active medium with Eq.(1.15) for the magneto-optically active medium, it is clear that both media exhibit circular birefringence, i.e., they are both gyrotropic. However, in contrast to the magneto-optical case, in media exhibiting natural optical activity the optical rotation does not depend on the direction of the propagation of the incident light. As a result, if a linearly polarized light beam traverses two times the same optical path in a chiral medium (e.g. a “round trip”), the emergent beam will be polarized in the same initial plane, in contrast to the Faraday effect. In other words, the natural optical activity is a reciprocal effect whereas the Faraday effect is not. However, it was experimentally observed that there are some special situations where the natural rotatory power also manifests a nonreciprocal character [26].

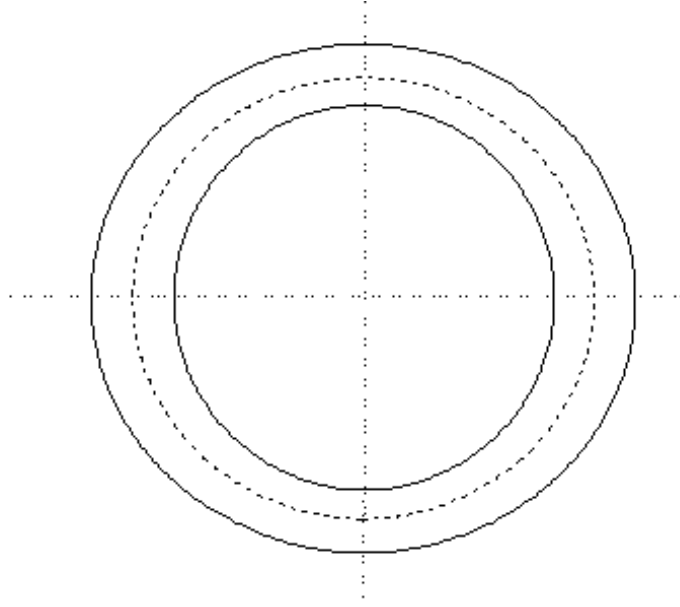


Figure 1-3: Dispersion law in chiral media in the frequency space. The dotted circle represents the case of an achiral isotropic media. The two full circles correspond to the two handedness of the chiral media (dextro- and laevo-rotatoria media).

1.3 Light propagation in homogeneous magnetochiral media

Although physically distinct, the Faraday effect and the natural optical rotatory power manifest themselves quite similarly as a rotation of the linear polarization of light. As discussed before, the first is associated with the breaking of the time-reversal symmetry by a magnetic field whereas the second is due to a lack of mirror symmetry in certain materials. This strong phenomenological resemblance between the Faraday effect and the optical rotatory power has motivated several works to search for a link between these two phenomena. The first attempt was made by Pasteur in 1848 who tried to generate handedness in crystals by applying an external magnetic field [27]. Motivated by the discovery of the Faraday effect two years before, Pasteur thought that a magnetic field could induce chirality in matter. This idea was later recognized as wrong by Lord Kelvin who stated that “the magnetic rotation has neither left-handed nor right-handed quality”. Indeed, the existence of a cross effect between the Faraday effect and the optical rotatory power is only allowed to occur under conditions where both mirror and time-reversal symmetries are *simultaneously* broken. Such an effect is called the

magnetochiral (MC) effect and was first predicted one century later by Groenewege in 1962 [28], followed by studies in crystalline materials [29]. The MC effect was later again predicted independently several times [30], [31], [32], [33], [34], both in absorption and in refraction, effects that are called magnetochiral dichroism and magnetochiral birefringence, respectively, following the terminology introduced first by Barron and Vrbancich [34].

In order to understand the MC effect in homogeneous and isotropic media, let us consider the propagation of light with wavevector $\mathbf{k}$ through a magneto-optically active chiral medium with dielectric tensor $\boldsymbol{\varepsilon}$ under the influence of a static magnetic field $\mathbf{B}$. Expanding $\boldsymbol{\varepsilon}$ to first order in $\mathbf{k}$ and $\mathbf{B}$, we have [29]:

$$\varepsilon_{ij}(\omega, \mathbf{k}, \mathbf{B}) = \varepsilon_0(\omega)\delta_{ij} + \alpha(\omega)i\epsilon_{ijl}k_l + \beta(\omega)i\epsilon_{ijl}B_l + \gamma(\omega)(\mathbf{B} \cdot \mathbf{k})\delta_{ij}, \quad (1.24)$$

where α and β are coefficients associated with the natural and magneto-optical activity, respectively. The last term of Eq. (1.24) describes the MC effect in homogeneous and isotropic media. It is important to point out some of the major features of this effect:

- i)* It depends on the relative orientation of $\mathbf{k}$ and $\mathbf{B}$.
- ii)* It has opposite sign for the two different chiral enantiomers (i.e., for the two mirror images of the medium).
- iii)* It is not circular differential, i.e., it does not depend on the state of polarization.

The estimated magnitude of the molecular MC effect is very weak (typically of the order of $\gamma Bk \approx 10^{-6}$ for $B \approx 1T$ at visible wavelengths), as expected for a cross-effect between the Faraday effect and the optical rotatory power, two already small effects. This probably explains why it has only been reported recently. The magnetochiral dichroism was first observed in 1997 by Rikken and Raupach [7]. They employed samples of diluted europium complexes $Eu((\pm)tc)_3$ (where $\pm$ indicates handedness), which exhibits the strongest chiral transition reported - the ${}^5D_0 \rightarrow {}^7F_{1,2}$ transition- to measure the difference in luminescence intensity in the direction parallel and antiparallel to the externally applied magnetic field. These measurements confirmed the existence of the MC effect in emission. The well-known relations between the Einstein coefficients in radiative processes [35] automatically suggest its existence in absorption, a suspicion that was experimentally observed one year later in measurements in samples of

$\alpha - NiSO_4 \cdot 6H_2O$ chiral crystals [36]. The measurements confirmed that the strength of the MC effect can be approximately estimated by the product of the strengths of the natural optical activity and magnetically induced optical activity [36].

In refraction, the first observation of the magnetochiral birefringence was reported by Klein-dienst and Wagnière [8], by performing interferometric measurements. Unfortunately, these measurements were about two orders of magnitude larger than the theoretical prediction [31]. This fact has motivated Vallet *et al.* to develop a new experimental setup to measure the MC birefringence [9]. Nevertheless, recent *ab initio* studies still reveal a significant discrepancy between theory and measurements of the MC in refraction [37].

The magnetochiral effect, specifically the magnetochiral dichroism, was also recently shown to favor the production of one enantiomer of a chiral molecule in a photochemical reaction, a process baptized enantioselective magnetochiral photochemistry [38]. In order to observe this, Rikken and Raupach have used the chiral complex Cr(III) tris-oxalato, which spontaneously dissociated and re-associates in solution, a process which is accelerated by the absorption of a photon [38]. In a solution of this complex at equilibrium there will always be right- and left-handed enantiomers in equal concentration, i.e., it will be a racemic mixture. The experiment concluded that in the presence of an unpolarized light beam propagating parallel to an externally applied magnetic field, a small excess of one enantiomer is produced. If the sense of the magnetic field was reversed, an equal amount of the mirror-image enantiomer was produced. This shows the presence of MC absorption.

The symmetry arguments which allow the existence of the MC effect in optics also apply to the case of the electrical transport. Indeed, it was recently demonstrated, both theoretically and experimentally for bismuth helices, that the electronical analogue of the optical MC effect exists [39]. In addition, recent experiments have established that charge transport through carbon nanotubes, which are molecular objects exhibiting chirality, are also subject to MC effects [40]. This experimental fact was supported by calculations using a free electron in a helix as a model of molecular chiral conductors [41], suggesting that MC anisotropy is a rather common phenomenon. Indeed, very recently Koerdt *et al.* have observed that light scattering in cholesteric liquid crystals exhibits MC anisotropy near the Bragg resonance [42]. All these recent studies show that MC effects are part of recent active research and have motivated the

theoretical work reported in this part of the thesis.

In the following we will discuss the existing theoretical models to describe the MC in homogeneous media.

1.3.1 Theory of the magnetochiral effect in homogeneous media

Classical model

The first attempt to understand the microscopic mechanism underlying the diamagnetic contribution to the MC effect is due to Baranova and Zel'dovich [31]. They have extended the already mentioned classical Becquerel model for magnetic optical activity and interpreted the MC effect as a consequence of Larmor precession in natural optical activity. In the presence of an external magnetic field $\mathbf{B}$, electrons in matter will undergo a Larmor precession with angular velocity with modulus $\Omega_L = -eB/2mc$, with m and e the electron mass and charge. Therefore LCP and RCP light will experience a dielectric constant at slightly shifted frequencies $\omega_{\pm} = \omega \pm \Omega_L$ from the incident light frequency ω :

$$\varepsilon(\omega) = \varepsilon(\omega \pm \Omega_L) = \varepsilon_0(\omega) \pm \frac{e}{2mc} \frac{\partial \varepsilon_0}{\partial \omega} B, \quad (1.25)$$

where we have assumed, for simplicity, that the magnetic field is parallel or antiparallel to the direction of light propagation. As a result, the coefficient $\beta(\omega)$ for the Faraday effect defined in Eq. (1.24) is given by:

$$\beta(\omega) = \frac{e}{2mc} \frac{\partial \varepsilon_0}{\partial \omega}. \quad (1.26)$$

If in addition the medium is composed of chiral molecules, one must also consider the second term in right-hand side of Eq. (1.24). Hence the effective dielectric constant experienced by the incident light can be written as:

$$\begin{aligned} \varepsilon(\omega, k, B, \sigma = \pm) &= \varepsilon_0(\omega \pm \Omega_L) \pm \alpha(\omega)(\omega \pm \Omega_L)k \\ &= \varepsilon_0(\omega) \pm \beta(\omega)B \pm \alpha(\omega)k + \frac{e}{2mc} \frac{\partial \alpha}{\partial \omega}(Bk). \end{aligned} \quad (1.27)$$

and one can identify the magnetochiral coefficient $\gamma(\omega)$ in Eq. (1.24) with

$$\gamma(\omega) = \frac{e}{2mc} \frac{\partial \alpha}{\partial \omega}. \quad (1.28)$$

Notice that for both polarization states the sign of the term proportional to Bk is the same, which confirms that the MC effect is polarization-independent. In spite of its simplicity, this classical model gives an insight about the physical mechanism governing the MC effect and it can even provide a correct order-of-magnitude estimate for the diamagnetic contribution to the MC dichroism [36].

Molecular quantum model

A detailed molecular theory of MC effect was developed by Wagnière [32],[33],[43],[44]. Within such an approach, the interaction between light and matter is treated quantum-mechanically and is described by the multipole interaction Hamiltonian

$$\mathcal{H}_{int} = -\boldsymbol{\mu} \cdot \mathbf{E}(t) - \mathbf{m} \cdot \mathbf{B}(t) - \mathbf{Q} : \nabla \mathbf{E}(t) + \dots, \quad (1.29)$$

with the electromagnetic field-operators given by

$$\begin{aligned} \mathbf{E}(t) &= \mathbf{E}_- \exp(-i\omega t) + \mathbf{E}_+ \exp(+i\omega t), \\ \mathbf{B}(t) &= \mathbf{B}_- \exp(-i\omega t) + \mathbf{B}_+ \exp(+i\omega t). \end{aligned} \quad (1.30)$$

In (1.29) $\boldsymbol{\mu}$ and $\mathbf{m}$ are the electric and magnetic dipole operators, $\mathbf{Q}$ is the electric quadrupole operator and so on. Although the electric dipole term dominates for the case of ordinary refraction and absorption (long-wavelength approximation), the description of the MC effect requires keeping the magnetic dipole and electric quadrupole terms as well.

In order to deduce the molecular MC effect in refraction, one should calculate the polarization $\mathbf{p}$ induced by the incident radiation in a molecule using quantum mechanics. Since the MC effect is a cross-effect between natural optical activity and magnetic optical activity, it is necessary to first calculate the contribution of these two optical effects to the molecular polarization $\mathbf{p}$.

The contribution $\mathbf{p}_{NOA}$ to $\mathbf{p}$ due to natural optical activity arises from both electric and magnetic dipole terms, though in non-isotropic media the electric quadrupole term may be also relevant. It is found that [44]:

$$\mathbf{p}_{NOA} = \sum_k \left\{ \frac{\langle i | \boldsymbol{\mu} | k \rangle \langle k | \mathbf{m} \cdot \mathbf{B}_- | i \rangle}{\hbar(\omega_{ki} - \omega)} + \frac{\langle i | \mathbf{m} \cdot \mathbf{B}_- | k \rangle \langle k | \boldsymbol{\mu} | i \rangle}{\hbar(\omega_{ki} + \omega)} \right\}, \quad (1.31)$$

which expresses the polarization induced by a photon with frequency ω exciting a molecule from its initial state $|i\rangle$ to a non-stationary superposition of states $|k\rangle$ and being re-emitted subsequently. In view of Eq. (1.31), notice that $\mathbf{p}_{NOA}$ contains products of electric transition moments $\langle i | \boldsymbol{\mu} | k \rangle$, parity-odd vectors, and magnetic transition moments $\langle k | \mathbf{m} | i \rangle$, parity-even vectors. Thus the corresponding optical activity tensor also contains products of the type $\boldsymbol{\mu}\mathbf{m}$, which is parity-odd. Natural optical activity will occur when the *product* of matrix elements of the type $\boldsymbol{\mu}\mathbf{m}$ is nonvanishing, which is the case of non-centrosymmetric homogeneous media or fluids of chiral molecules.

For magnetic optical activity one has also to consider the interaction of the molecular magnetic dipole with the external magnetic field $\mathbf{B}_{ext}$. Therefore the contribution to the molecular polarization is of the form [44]:

$$\begin{aligned} \mathbf{p}_{MOA} = & \sum_k \sum_n \frac{\langle i | \boldsymbol{\mu} | n \rangle \langle n | \boldsymbol{\mu} \cdot \mathbf{E}_- | k \rangle \langle k | \mathbf{m} \cdot \mathbf{B}_{ext} | i \rangle}{\hbar^2 (\omega_{ni} - \omega) \omega_{ki}} \\ & + \text{similar terms.} \end{aligned} \quad (1.32)$$

Notice that (1.32) involves a product of the type $\boldsymbol{\mu}\boldsymbol{\mu}\mathbf{m}$, a parity-even quantity, which is consistent with the symmetry requirement that the polarization vector must be even upon time reversal. The corresponding optical activity tensor will also contain a parity-even product of the type $\boldsymbol{\mu}\boldsymbol{\mu}\mathbf{m}$ which therefore implies that magnetic optical activity can occur in all materials.

In order to generate the MC effect in refraction one has to take into account a combination of the magnetic dipole interaction with $\mathbf{B}_{ext}$ and a magnetic dipole, or an electric quadrupole interaction with the light field. The MC contribution to the molecular polarization has the

form:

$$\begin{aligned} \mathbf{p}_{MC} = & \sum_k \sum_n \frac{\langle i | \boldsymbol{\mu} | n \rangle \langle n | \mathbf{m} \cdot \mathbf{B}_- | k \rangle \langle k | \mathbf{m} \cdot \mathbf{B}_{ext} | i \rangle}{\hbar^2 (\omega_{ni} - \omega) \omega_{ki}} \\ & + \text{similar terms}, \end{aligned} \quad (1.33)$$

which contains parity-odd products of the kind $\boldsymbol{\mu}\mathbf{m}\mathbf{m}$. Consequently, the MC effect is expected to occur only in chiral media subject to a magnetic field. Furthermore, it can be shown that any effect involving an odd number of magnetic dipole and/or electric quadrupole interactions will depend on the state of polarization of the electromagnetic wave (the so-called circular-differential effects) [32]. Thus the MC effect is not circular-differential.

The description of the MC effect in absorption or refraction is similar but in this case the relevant quantity to be evaluated is the transition probability per unit of time from the initial state $|i\rangle$ to the final one $|f\rangle$:

$$W_{i \rightarrow f}(\omega) = \frac{2\pi}{\hbar^2} |\langle i | \mathcal{H}_{int}(\omega) | f \rangle|^2 \delta(\omega_{fi} - \omega). \quad (1.34)$$

The interaction between the incident radiation and the molecular system is again described by the interaction Hamiltonian (1.29) that, when inserted into Eq. (1.34), will generate several terms. Among them, one can distinguish the electric dipole-magnetic dipole and the electric dipole-electric quadrupole terms, which are parity-odd and associated with the circular dichroism in chiral media. In the presence of an external magnetic field $\mathbf{B}_{ext}$ parallel to the direction of the incident or emitted radiation, the initial and final molecular states will be perturbed. The resulting electric dipole-electric dipole term will be related to magnetic circular dichroism whereas the electric dipole-magnetic dipole and the electric dipole-electric quadrupole terms will contribute to the MC dichroism. Alternatively, one can describe MC dichroism as well as related optical phenomena in absorption and emission by considering damping in the process of molecular polarization [34]. As a result, the molecular susceptibility will be complex-valued and its imaginary part is associated with the dichroism.

It is important to recall that these theoretical quantum models of the MC effect apply only to *one* molecule with (average) polarization vector $\mathbf{p}$. Their applicability to a homogeneous medium composed of N molecules per unit volume can be realized by considering the Clausius-

Mossotti equation which, in an isotropic medium, reads [11]:

$$\frac{\varepsilon - 1}{\varepsilon + 2} = \frac{4\pi N}{3} \gamma_{mol}, \quad (1.35)$$

which relates the macroscopic dielectric constant ε to the microscopic molecular polarizability $\gamma_{mol} = \mathbf{p}/\mathbf{E}$. In Chapter 4 we will introduce an alternative model to describe how MC effects could be generated in *inhomogeneous light scattering media*.

1.3.2 Magnetochiral effect: an extraterrestrial source of handedness in life?

The fact that the magnetochiral effect acts as an enantioselective mechanism may provide a possible explanation for one of the most fundamental and fascinating topics in science: the origin of the homochirality of life and, consequently, of life itself [43]. It is well-known that living beings use only the left-handed enantiomers of aminoacids in order to synthesize proteins. Although an entire “mirror-image biology” using only right-handed aminoacids is in principle possible, nobody knows exactly why nature works only in a “left-handed” mode. There are some theories developed to answer this question. One of them is the parity-violating electroweak interaction between the aminoacids which is, however, predicted to be extremely small ($\sim 10^{-17}$) and probably largely insufficient to induce the observed enantiomeric excess. Another possible answer is based on photochemistry with circularly polarized light. However, the small amount of natural circularly polarized light available on Earth has led to the speculation that homochirality could have an extraterrestrial origin. This hypothesis is supported by two recent discoveries: an excess of left-handed aminoacids on meteorites [45], suggesting that homochirality was somehow “seeded” by the impact of one of these materials on Earth, and that of significant amounts of circularly polarized light in astronomical observations [46]. Within this context, the hypothesis of enantioselective magnetochiral photochemistry as an explanation of the origin of homochirality of life fits quite well since high magnetic fields and unpolarized light are more common in universe than circularly polarized light. At the time of writing, it is not precisely known how large an excess of left-handed aminoacids is required in order to induce biological homochirality and if the magnetochiral effect can indeed be held responsible for homochirality given the natural magnetic fields. Some authors argue that enantioselective

magnetochiral photochemistry cannot solve the problem of homochirality of life in view of the weakness of the effect [47]. There are thus several points to be clarified, showing that the problem of the origin of the homochirality of life is far from being solved.

Résumé du Chapitre 1 -

Propagation de la lumière dans les milieux homogènes chiraux et/ou magnéto-optiques

Ce chapitre est consacré à une brève description de la propagation de la lumière dans les milieux homogènes avec des symétries brisées : milieux magnéto-optiques (où la symétrie du renversement temporel est brisée), milieux chiraux (où la symétrie de parité est brisée) et les milieux magnéto-chiraux (où les symétries de parité et du renversement du temps sont simultanément brisées).

Propagation de la lumière dans les milieux magnéto-optiques

La présence d'un champ magnétique externe $\mathbf{B}$, responsable pour la brisure de la symétrie du renversement du temps, modifie les relations constitutives des milieux optiques. Pour les fréquences optiques, l'influence de $\mathbf{B}$ dans le tenseur diélectrique ε est faible et peut être donc traitée comme une perturbation :

$$\varepsilon_{ij}(\mathbf{B}) = \varepsilon_0 \delta_{ij} + \varepsilon_F \Phi_{ij}, \quad (1.36)$$

où $\varepsilon_0 \equiv n_0^2$ est la constante diélectrique à l'absence de $\mathbf{B}$, le tenseur antisymétrique Φ est défini comme $\Phi_{ij} \equiv i\epsilon_{ijk}\hat{B}_k$ et le paramètre sans dimension ε_F est donné par :

$$\varepsilon_F = 2n_0Vc_0B/\omega, \quad (1.37)$$

où V est la constante de Verdet.

Il est important de signaler que ε dans les milieux magnéto-optiques possède des relations de symétrie très particulières. Des arguments de symétrie imposent que ε obéisse les relations suivantes [12]:

$$\varepsilon_{ij}(\mathbf{B}) = \varepsilon_{ji}(-\mathbf{B}) \neq \varepsilon_{ji}(\mathbf{B}), \quad (1.38)$$

qui sont les équivalents optiques des relations d'Onsager en thermodynamique [13]. La magnéto-optique étudie la manière dont la propagation de la lumière est modifiée par $\mathbf{B}$ via Eq. (1.36) et les nouveaux phénomènes que cette modification implique. Le plus connu des effets magnéto-optiques est *l'effet Faraday* que sera traité au paragraphe suivant.

L'effet Faraday

L'origine de l'effet Faraday (ou activité optique magnétiquement induite) peut être comprise en déduisant la relation de dispersion dans les milieux magnéto-optiques. Elle possède la forme :

$$\omega(k) = \frac{kc_0}{n_0} + \frac{\varepsilon_F c_0}{2n_0^3} \sigma(\hat{\mathbf{k}}) \hat{\mathbf{B}} \cdot \mathbf{k}, \quad (1.39)$$

où $\sigma(\hat{\mathbf{k}}) = \pm 1$ désigne les états de polarisation circulaire de la lumière incidente. La relation de dispersion (1.39) implique que la surface à fréquence constante soit décalée d'une quantité $\varepsilon_F c_0 / 2n_0^3$ à partir de l'origine. Le décalage se produit dans le même sens, ou dans le sens opposé à $\mathbf{B}$, en dépendant de l'état de la polarisation incidente, comme illustre la figure (1-2). Comme conséquence, pour un faisceau incident linéairement polarisé, le vecteur de polarisation émergent subira une rotation $\alpha = VBL$ selon la direction de $\mathbf{B}$, où L est le chemin optique parcouru. Remarquons que la rotation Faraday change de signe si le sens de $\mathbf{B}$ relatif à celui de la propagation de la lumière change et que l'angle α est multiplié par deux dans un chemin

d'aller-retour de la lumière dans le milieu.

Il exist d'autres effets magnéto-optiques importants, comme l'effet Kerr et Cotton-Mouton et la biréfringence magnétoélectrique [18] et la biréfringence magnétoélectrique de Jones [21], ces deux dernières ayant été observées plus récemment.

Propagation de la lumière dans les milieux chiraux

La propagation de lumière dans les milieux qui ne possèdent pas la symétrie miroir génère le phénomène de l'activité optique naturelle (AON). Il est important de remarquer que l'AON constitue un autre exemple de *gyrotropie*, i.e., la propriété que certains milieux ont de distinguer la lumière circulairement polarisée à gauche de la lumière circulairement polarisée à droite. La gyrotropie peut également être induite par l'application d'un champ magnétique externe, comme c'est le cas de l'effet Faraday. Nous pouvons donc procéder de façon analogue au traitement de l'effet Faraday, discutée au paragraphe précédent, et dériver la relation de dispersion dans les milieux chiraux :

$$\omega(k) = \frac{kc_0}{n_0}(1 \pm f), \quad (1.40)$$

où f est un paramètre qui caractérise la magnitude de l'AON et le signe $\pm$ est associé à la « handedness » du milieu chiral. La relation de dispersion (1.40) est représentée à la figure 1-3 qui montre son caractère isotrope, en contraste avec la relation de dispersion de l'effet Faraday (1.39). La comparaison entre les relations de dispersion (1.39) et (1.40) révèle encore que l'AON ne dépend pas de la direction de la lumière incidente, contrairement à l'effet Faraday. En conséquence, une onde linéairement polarisée qui traverse le même chemin optique à deux reprises (e.g., un « aller-retour ») dans un milieu chiral sortira polarisée dans le même plan initial, différemment de l'effet Faraday. Autrement dit, l'AON est un effet réciproque alors que l'effet Faraday n'est pas.

Propagation de la lumière dans les milieux magnéto-chiraux

L'effet magnéto-chiral peut être vu comme un effet hybride entre l'effet de l'activité optique naturelle et l'effet de l'activité optique magnétiquement induite. Il a été prédit théoriquement pour la première fois en 1962 par Groenewege [28] (puis indépendamment en [29][30][31][32][33][34]) mais observé récemment seulement, en absorption [7] et en refraction [8][9], car il s'agit d'un effet particulièrement faible (typiquement de l'ordre de 10^{-6}).

Afin de déduire l'effet magnéto-chiral dans les milieux homogènes, considérons la propagation de la lumière (vecteur d'onde $\mathbf{k}$) dans un milieu magnéto-optique et chiral (tenseur diélectrique ϵ) sous un champ magnétique externe $\mathbf{B}$. L'expansion de ϵ jusqu'à première ordre en $\mathbf{B}$ et $\mathbf{k}$ nous fournit [29] :

$$\epsilon_{ij}(\omega, \mathbf{k}, \mathbf{B}) = \epsilon(\omega)\delta_{ij} + \alpha(\omega)i\epsilon_{ijl}k_l + \beta(\omega)i\epsilon_{ijl}B_l + \gamma(\omega)(\mathbf{B} \cdot \mathbf{k})\delta_{ij}, \quad (1.41)$$

où α et β sont des coefficients associés, respectivement, à l'activité optique naturelle et l'activité optique magnétiquement induite. Le dernier terme en (1.41) décrit l'effet magnéto-chiral dans les milieux homogènes et isotropes. Il est important de remarquer les caractéristiques principales de cet effet :

- i)* il ne dépend pas de l'orientation relative entre $\mathbf{k}$ et $\mathbf{B}$.
- ii)* il change de signe pour les deux énantiomères chiraux (i.e., les deux images miroir du milieu).
- iii)* il ne dépend pas de l'état de polarisation de la lumière.

De récentes expériences conduites à Grenoble ont montré que l'effet magnéto-chiral est capable de produire un excès d'un des énantiomères dans une réaction photochimique [38]. Ce fait a stimulé le débat sur les possibles implications de l'effet magnéto-chiral à l'origine de l'homochiralité de la vie [43], un des problèmes ouverts majeurs en science.

Dans le chapitre 4 nous allons nous intéresser à l'étude de l'effet magnéto-chiral dans les milieux inhomogènes.

Chapter 2

Microscopic single scattering by point particles

This chapter is devoted to the study of single scattering of vector waves by *point particles*, i.e., scatterers with a typical size much smaller than the wavelength of the incident radiation. This approximation will be used throughout this thesis and is extremely useful since it considerably simplifies the treatment of multiple scattering. The consideration of the exact Mie solution for single scattering by an arbitrary object makes the treatment of multiple scattering difficult and cumbersome, and does not always lead to new physics. Although simpler than Mie scattering, the microscopic approach to treat wave scattering by pointlike particles still presents some difficulties, such as the appearance of singularities in Green's functions that describe the propagation of the scattered wave. This problem is, however, conveniently treated by using regularization procedures, as will be discussed in the following.

The use of pointlike scatterers is valuable not only because it simplifies the treatment of multiple scattering, but also because it adapts very well to some experimental situations. Point scatterers can represent finite-size systems with an internal degree of freedom such as two-level atoms exhibiting one resonance in solid-state or in atomic physics. For a detailed description of the point-dipole model, including several applications such as the calculation of the local density of states of electromagnetic waves, the reader is referred to Ref. [48].

We will introduce the Green's function formalism and the concept of the T-matrix. The

T-matrix for single scattering relates the incident and scattered wave and thus constitutes the building block of multiple scattering. It contains all information about the scattering process (polarization, cross-sections, time delay, etc.) and exhibits the underlying symmetry principles.

2.1 Green's function formalism

In a scalar treatment for electromagnetic waves, the Green function G_0 is defined as the solution of the inhomogeneous scalar wave equation:

$$\left[\left(\frac{\omega}{c_0} + i\epsilon \right)^2 + \nabla^2 \right] G_0(\omega, \mathbf{r} - \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'). \quad (2.1)$$

Notice that we consider translationally invariant media, $G_0(\mathbf{r}, \mathbf{r}') = G_0(\mathbf{r} - \mathbf{r}')$. Physically, Eq. (2.1) describes wave propagation from an external δ -function source located at $\mathbf{r}'$ to the observation point $\mathbf{r}$. An infinitesimal positive imaginary part $i\epsilon$ was added to the frequency ω in order to guarantee the correct analytical properties of G_0 in the complex plane. The explicit forms of G_0 in 3D Fourier and coordinate space are:

$$G_0(\omega, \mathbf{p}) = \frac{1}{k^2 - p^2 + i\epsilon}, \quad (2.2)$$

$$G_0(\omega, \mathbf{r}) = -\frac{\exp(ikr)}{4\pi r}. \quad (2.3)$$

If one wishes to take into account the polarization of electromagnetic waves, it is necessary to consider the Helmholtz equation:

$$\left[\left(\frac{\omega}{c_0} + i\epsilon \right)^2 \mathbf{I} - \nabla \times \nabla \times \right] \mathbf{G}_0(\omega, \mathbf{r} - \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \mathbf{I}, \quad (2.4)$$

which involves now the Green tensor $\mathbf{G}_0$ and $\mathbf{I}$ is the unit tensor. By Fourier transforming Eq. (2.4), we obtain

$$\begin{aligned} \mathbf{G}_0(\omega, \mathbf{p}) &= \frac{1}{(k^2 + i\epsilon)\mathbf{I} - p^2 \Delta_{\mathbf{p}}} = \frac{1}{k^2 - p^2 + i\epsilon} \Delta_{\mathbf{p}} + \frac{1}{k^2} \hat{\mathbf{p}} \hat{\mathbf{p}} \\ &\equiv \mathbf{G}_0^T(\omega, \mathbf{p}) + \mathbf{G}_0^L(\omega, \mathbf{p}), \end{aligned} \quad (2.5)$$

where $(\Delta_{\mathbf{p}})_{ij} \equiv \delta_{ij} - p_i p_j / p^2$ is the projector upon the space of transverse fields (normal to $\mathbf{p}$). In Eq. (2.5) we have performed a decomposition of the Green tensor into a transverse (T) and a longitudinal (L) part with respect to the momentum $\mathbf{p}$. The transverse part is associated with propagating modes whereas the longitudinal part describes (electrostatic) modes that do not propagate. For this reason the longitudinal part is generally neglected. However, it contributes to the evaluation of the local field and can be important to describe polarization properties in multiple scattering [49].

In real space, the Green tensor is given by [48]:

$$\mathbf{G}_0(\omega, \mathbf{r}) = -\frac{\exp(ikr)}{4\pi r} [P(ikr)\mathbf{I} + Q(ikr)\widehat{\mathbf{r}}\widehat{\mathbf{r}}] + \frac{\delta(\mathbf{r})}{3k^2}\mathbf{I}, \quad (2.6)$$

with the functions P and Q defined according to

$$P(x) \equiv \left(1 - \frac{1}{x} + \frac{1}{x^2}\right), \quad Q(x) \equiv \left(-1 + \frac{3}{x} - \frac{3}{x^2}\right). \quad (2.7)$$

The $\delta(\mathbf{r})/3k^2$ term in (2.6) ensures that the average field inside a spherical region containing the dipole is nonzero and is related to the Lorentz local field in electrostatics [11].

Equation (2.6) can be decomposed in a transverse and longitudinal part [48]:

$$\mathbf{G}_0^T(\omega, \mathbf{r}) = -\frac{\mathbf{I} - 3\widehat{\mathbf{r}}\widehat{\mathbf{r}}}{4\pi k^2 r^3} - \frac{\exp(ikr)}{4\pi r} [P(ikr)\mathbf{I} + Q(ikr)\widehat{\mathbf{r}}\widehat{\mathbf{r}}]; \quad \mathbf{G}_0^L(\omega, \mathbf{r}) = \frac{\mathbf{I} - 3\widehat{\mathbf{r}}\widehat{\mathbf{r}}}{4\pi k^2 r^3} + \frac{\delta(\mathbf{r})}{3k^2}\mathbf{I} \quad (2.8)$$

Both parts of the $\mathbf{G}_0$ exhibit singularities at $r = 0$. In order to remove these divergencies we will appeal to regularization procedures, as will be discussed in the next section.

2.2 Scattering from point particles: T-matrix

In this section we define and formulate the T-matrix of a single point scatterer of vector waves. In order to accomplish this, one should introduce an external source in the right-hand side of the Helmholtz equation (2.4) which, in real space, reads

$$\left[\left(\frac{\omega}{c_0} + i\epsilon \right)^2 \mathbf{I} - \nabla \times \nabla \times \right] \mathbf{G}(\omega, \mathbf{r}, \mathbf{r}') = \mathbf{I} \delta(\mathbf{r} - \mathbf{r}') - \frac{\omega^2}{c_0^2} \chi(\mathbf{r}) \mathbf{G}(\omega, \mathbf{r}, \mathbf{r}'), \quad (2.9)$$

where $\chi(\mathbf{r})$ is the dimensionless electric susceptibility of the point particle. It is related to the dielectric tensor according to $\chi(\mathbf{r}) \equiv \varepsilon(\mathbf{r}) - 1$. Introducing the frequency-dependent potential operator $\mathbf{V}(\omega) \equiv -(\omega/c_0)^2 \chi$ [49], Eq. (2.9) can be written as

$$\mathbf{G}(\omega) = \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{G}(\omega). \quad (2.10)$$

Equation (2.10) is called the Lippmann-Schwinger equation and constitutes the basis for the theory of multiple scattering. The potential associated with a pointlike particle located at the origin $\mathbf{r} = 0$ has the following matricial form in real space:

$$\mathbf{V}(\omega, \mathbf{r}) \equiv -(\omega/c_0)^2 \alpha_B \delta(\mathbf{r}) \mathbf{I}, \quad (2.11)$$

where α_B is the bare polarizability of the scatterer, which measures the strength of the coupling between the electromagnetic waves and the particle. It has the dimension of a volume which should relate to the physical size of the scatterer. Hence, the typical linear dimension of the point scatterer, $a \ll \lambda$, is $a \approx \alpha_B^{1/3}$. For an atom, a is typically of the order of the Bohr radius. The bare polarizability α_B differs from the static polarizability $\alpha(0)$, which is actually measured in a experiment. This difference arises from the fact α_B does not take into account depolarization effects that occur at the boundary of the scatterer.

The scattering T-matrix $\mathbf{T}(\omega)$ is defined by the equation

$$\mathbf{G}(\omega) \equiv \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{G}_0(\omega), \quad (2.12)$$

and can explicitly be obtained by iterating $\mathbf{G}_0$ in the integral equation (2.10). This results in the so-called *Born series*:

$$\begin{aligned} \mathbf{T}(\omega) &= \mathbf{V}(\omega) + \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) + \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) + \dots \\ &\equiv t(\omega) \delta(\mathbf{r}) \mathbf{I}, \end{aligned} \quad (2.13)$$

where the T-matrix element $t(\omega)$ has the form

$$t(\omega) = -(\omega/c_0)^2 \frac{\alpha_B}{1 + (\omega/c_0)^2 \alpha_B G_0(\omega, r=0)}. \quad (2.14)$$

The T-matrix is related to the frequency-dependent polarizability $\alpha(\omega)$ according to:

$$\alpha(\omega) = -t(\omega)/(\omega/c_0)^2. \quad (2.15)$$

Physically, $\alpha(\omega)$ represents the volume of the point particle actually “seen” by the incoming light wave. The total T-matrix $\mathbf{T}_{tot}(\omega, \mathbf{r}, \mathbf{r}')$,

$$\mathbf{T}_{tot}(\omega, \mathbf{r}, \mathbf{r}') = t(\omega) \delta(\mathbf{r} - \mathbf{R}) \delta(\mathbf{r}' - \mathbf{R}) \mathbf{I}, \quad (2.16)$$

connects the scattered electric field $\mathbf{E}_{sca}(\omega, \mathbf{r}')$ at the position $\mathbf{r}'$ from a point particle at the position $\mathbf{R}$ and the incident field $\mathbf{E}_{inc}(\omega, \mathbf{r})$ at $\mathbf{r}$:

$$\mathbf{E}_{sca}(\omega, \mathbf{r}') = \mathbf{T}_{tot}(\omega, \mathbf{r}, \mathbf{r}') \cdot \mathbf{E}_{inc}(\omega, \mathbf{r}). \quad (2.17)$$

Using Eq. (2.12), we obtain the corresponding Green’s function

$$\mathbf{G}(\omega, \mathbf{r}, \mathbf{r}') \equiv \mathbf{G}_0(\omega, \mathbf{r} - \mathbf{r}') + t(\omega) \mathbf{G}_0(\omega, \mathbf{R} - \mathbf{r}) \cdot \mathbf{G}_0(\omega, \mathbf{r}' - \mathbf{R}). \quad (2.18)$$

The first term in Eq. (2.18) represents the unscattered wave from $\mathbf{r}'$ to $\mathbf{r}$. The second term describes the wave that propagates from $\mathbf{r}'$ to the position $\mathbf{R}$ of the point particle and subsequently propagates to the point $\mathbf{r}$, after being scattered by the particle.

It is important to point out the T-matrix (2.16) contains all information about the scattering process, not only about its “far field” properties. For this reason, it is also called “off-shell” T-matrix. In the far-field regime ($kr \gg 1$), Eq. (2.16) can be considerably simplified [48]. In this regime, the T-matrix is called “on-shell” and obviously contains much less information than the “off-shell” form, though sufficient to find the scattering cross-section.

As can be seen from Eqs. (2.6) and (2.14), the Green tensor $\mathbf{G}_0$ exhibits a singularity at $\mathbf{r} = 0$, resulting in a nonphysical zero T-matrix. In order to treat this problem, we shall employ

regularization techniques. The longitudinal $\overline{G}_0^L$ and transverse $\overline{G}_0^T$ Green tensor at $r = 0$ are regularized according to [48]:

$$\overline{G}_0^T(\omega, r = 0) \rightarrow -\left(\frac{\Lambda_T}{6\pi} + i\frac{k}{6\pi}\right), \quad (2.19)$$

$$\overline{G}_0^L(\omega, r = 0) \rightarrow \frac{\Lambda_L^3}{6\pi k^2}, \quad (2.20)$$

where Λ_T and Λ_L are inverse length scales that play the role of a momentum cut-off in the regularization process. Their physical significance will become clear in the following. Inserting the finite values of the Green tensor $\overline{\mathbf{G}}_0^L$ and $\overline{\mathbf{G}}_0^T$ [Eqs. (2.19) and (2.20)] into Eq. (2.14), we obtain the expression for the regularized T-matrix for point scatterers:

$$t(\omega) = -\frac{(\omega/c_0)^2}{\alpha_B^{-1} + \Lambda_L^3/6\pi - (\omega/c_0)^2\Lambda_T/6\pi - i(\omega/c_0)^3/6\pi}. \quad (2.21)$$

We can now relate the parameters α_B , Λ_L and Λ_T to real physical quantities. Using Eq. (2.15), it is possible to relate the parameters α_B and Λ_L to the frequency-independent or static polarizability $\alpha(0)$:

$$\alpha(0) = \frac{\alpha_B}{1 + \alpha_B\Lambda_L^3/6\pi}. \quad (2.22)$$

The transverse regularization parameter Λ_T is related to the resonance frequency ω_0 and to the linewidth Γ of the resonant T-matrix:

$$(\omega_0/c_0)^2 \equiv \frac{6\pi}{\Lambda_T\alpha(0)}, \quad \Gamma = \frac{\omega_0^2}{c_0\Lambda_T}. \quad (2.23)$$

It is also useful to introduce the quality factor $Q \equiv \omega_0/\Gamma$ of the resonance, and to relate it to Λ_T according to $\Lambda_T = \omega_0 Q/c_0$. This shows that the regularization parameters Λ_L and Λ_T are not mere mathematical tools introduced to get rid off infinities in the Green tensor, but they also have a physical interpretation. Notice that Λ_L and Λ_T are related to static and dynamic quantities, respectively. Equations (2.22) and (2.23) define the set of experimentally accessible parameters [ω_0 , Γ and $\alpha(0)$] that characterize the point scatterer¹. Finally, Λ_L can be related

¹Notice that only two of them are independent.

to the size of the particle. Consequently, if the volume $\Lambda_L^3 \approx V_0^{-1}$ of the scatterer is known, it is possible to estimate the bare polarizability α_B by measuring $\alpha(0)$. For an atom, for example, Λ_L^{-1} is typically equal to its Bohr radius, which can be regarded as the “size” of the electronic wave function.

The final expression for the T-matrix of a point scatterer in terms of the physical parameters describing a classical two-level system is thus given by:

$$t(\omega) = -(\omega/c_0)^2 \alpha(0) \frac{\omega_0^2}{\omega_0^2 - \omega^2 - i(\Gamma\omega^3/\omega_0^2)}. \quad (2.24)$$

From this expression it is clear that the imaginary part of the T-matrix is related to the linewidth of the resonance, showing that the finite width of the resonance is due to radiative damping.

The T-matrix obeys the fundamental Optical Theorem [50]:

$$-\frac{\text{Im } t(\omega)}{k} = \frac{|t(\omega)|^2}{6\pi} = \sigma_{sca}(\omega). \quad (2.25)$$

The optical theorem expresses the conservation of energy during the scattering process: the energy removed from the incident wave (or extinction, represented by the left-hand side of Eq. (2.25)) is entirely converted into the scattered intensity given by σ_{sca} , provided there is no absorption. At resonance $\omega = \omega_0 = 2\pi c_0/\lambda_0$, the scattering cross section reaches its maximal value

$$\sigma_{sca}(\omega_0) = \frac{3}{2} \frac{\lambda_0^2}{\pi}, \quad (2.26)$$

which reflects the unitarity of the $\mathbf{S}$ matrix, $\mathbf{S} = \mathbf{I} - 2\pi i \mathbf{T}$ [50].

2.2.1 Applications of the point dipole model

In this section we will treat four physical objects that can be appropriately described by the dipole model discussed in the previous section: a small dielectric sphere, an atom, a magneto-optical active scatterer and a chiral molecule.

Small dielectric sphere

In order to calculate the polarizability of a small dielectric sphere of volume $V = 4\pi a^3/3$ and dielectric constant ε , it is well-known that depolarization effects at the boundaries of the sphere have to be taken into account [11]. For this reason, the experimentally accessible quantity in electrostatics is not the bare polarizability $\alpha_B = (\varepsilon - 1)V$, but rather the static polarizability

$$\alpha(0) = 3 \frac{\varepsilon - 1}{\varepsilon + 2} V. \quad (2.27)$$

These depolarization effects also play an important role in the derivation of the Clausius-Mossotti relation [11]. Comparing Eqs. (2.27) and (2.22), we obtain the relation $\Lambda_L^3/6\pi = 1/3V$ that shows that the cut-off parameter Λ_L^{-1} can be related to the size of the pointlike scatterer, confirming the assertion made earlier that it represents a geometrical depolarization effect. For small dielectric spheres the resonances are geometrical, in contrast to atoms whose resonances are due to internal degrees of freedom.

The expression (2.24) for the T-matrix of a pointlike scatterer can also be derived from the exact Mie T-matrix t_{Mie} corresponding to a sphere of arbitrary size (for standard books on Mie scattering, see Refs. [51][52][53]) in certain cases. In the limit where the typical size a of the scatterer is much smaller than wavelength of the radiation inside and outside the scatterer (i.e., $ka \ll 1$ and $\sqrt{\varepsilon}ka \ll 1$), called the *Rayleigh limit*, the Mie solution can be written as $t_{Mie} \simeq -k^2\alpha(0)$ with $\alpha(0)$ given by (2.27). The cross section of a Rayleigh scatterer vanishes as ω^4 , a behavior that explains the blue color of the sky. In addition, the Rayleigh differential cross section $d\sigma_{scat}(\omega)/d\Omega$, which describes the angular distribution of the scattered intensity, has the form [49]:

$$\frac{d\sigma_{scat}(\omega)}{d\Omega} = \frac{1}{2} \frac{1}{(4\pi)^2} |t(\omega)|^2 (1 + \cos^2 \theta). \quad (2.28)$$

No resonance effects occur in the Rayleigh limit. However, when $ka \ll 1$ but $\sqrt{\varepsilon}ka \gg 1$, resonant effects become important [51].

Lorentz model for an atom

The Lorentz model for an atom subject to a static electric field consists of an electron (charge e and mass m) elastically bound to the nucleus at a distance $\mathbf{r}$ from the center of mass of the atom. The electron is treated thus as a classical damped oscillator with resonant frequency ω_0 and a damping force proportional to Γ . Within this model, the regularization parameter Λ_T can be identified with the classical radius of the electron: using Eq. (2.23) and the expression for the radiative damping within the Lorentz model, $\Gamma = \omega_0^2 e^2 / 6\pi\epsilon_0 m c_0^3$, one identifies

$$\Lambda_T = \frac{3}{2r_0}, \quad (2.29)$$

where $r_0 = e^2 / 4\pi\epsilon_0 m c_0^2$ is the classical electron radius. The relation of Λ_T to r_0 seems to be natural within a classical model since r_0 is the only length scale in this problem that can be constructed with only classical constants [49].

Pointlike scatterer in a magnetic field

Another important application of the pointlike scatterer model is the description of the scattering properties of a magneto-optical pointlike scatterer. Since the pointlike scatterer can appropriately model a two-level atom, the application of an external magnetic field $\mathbf{B}$ will induce the Zeeman effect. Using a similar approach to those described in the previous section, van Tiggelen, Maynard and Nieuwenhuizen have obtained the T-matrix of a pointlike scatterer under the influence of an external magnetic field [54]. Using Eq. (1.11) for the dielectric tensor in a magnetic field they have showed that, up to linear order in $\mathbf{B}$, the T-matrix has the form

$$\mathbf{T}(\mathbf{B}, \omega) = t_0 + t_1 \Phi, \quad (2.30)$$

with the coefficients t_0 and t_1 given by:

$$t_0 = -\frac{4\pi(\epsilon_0 - 1)x^3}{\omega(\epsilon_0 + 2)}, \quad t_1 = -\frac{12\pi\epsilon_F x^3}{\omega(\epsilon_0 + 2)^2}, \quad (2.31)$$

where $x = ka$ is the size parameter of the scatterer.

Chiral molecule composed of point particles

Another possible application is to model molecular systems and, in particular, chiral molecules as N point particles. As it will be discussed in Chapter 4, a chiral object must be composed of at least four point particles. This constitutes the simplest model for chiral molecules. Using this simple model, we will investigate in Chapters 4 and 5 light scattering in chiral and magnetochiral random media.

2.2.2 T-matrices and symmetry principles

The symmetries laws that govern electromagnetic propagation manifest themselves in the T-matrix in a crucial way. The T-matrix exhibits the symmetries associated with the scatterers, such as parity, rotation, time-reversal, etc., as well as their constitutive relations discussed in the previous chapter. In fact, it is possible to derive, using group theory, the form of the T-matrix only by looking at the symmetry properties of the scatterer or, more precisely, by analyzing symmetry groups associated with the scatterer [55].

The impact of symmetry laws on scattering becomes more evident if the T-matrix is expressed in the polarization basis defined by Van-de-Hulst [51]. This basis is defined by two vectors, one lying in the so-called scattering plane (the plan containing $\mathbf{k}$ and $\mathbf{k}'$) and the other orthogonal to it. With σ and σ' the helicity² of incident and scattered waves, respectively, the T-matrix in the Van-the-Hulst basis and in the presence of an external magnetic field $\mathbf{B}$ is denoted by:

$$T_{\mathbf{k}\sigma, \mathbf{k}'\sigma'}(\mathbf{B}) = \langle \sigma(\mathbf{k}), \mathbf{k} | \mathbf{T}(\mathbf{B}, \omega) | \sigma(\mathbf{k}'), \mathbf{k}' \rangle . \quad (2.32)$$

A *parity* operation is equivalent to a spatial inversion. If the scatterer is symmetric under a mirror operation, its T-matrix must obey the parity relation:

$$\text{Parity:} \quad T_{-\mathbf{k}\sigma, -\mathbf{k}'\sigma'}(\mathbf{B}) = T_{\mathbf{k}\sigma, \mathbf{k}'\sigma'}(\mathbf{B}) \quad (2.33)$$

A *chiral* scatterer is not invariant under a mirror operation and consequently *does not* verify

²The helicity values $\sigma(\mathbf{k}) = \pm 1$ correspond, respectively, to right and left circularly polarized light.

Eq. (2.33). On the other hand, an achiral *magneto-optical* scatterer *does* obey it.

The *reciprocity principle* plays an important role in scattering theory. The concept of reciprocity can be expressed by the statement: “If I can see you, you can see me”. In scattering language, this means that upon interchanging the positions of source and detector, the scattering properties will remain the same. It is important to emphasize that the reciprocity principle does not rely uniquely on the properties of the scatterer, as is true for parity, but also on the form of Maxwell equations. A rigorous proof of the reciprocity principle, derived from the Maxwell equations, can be found in Ref. [12].

The reciprocity principle implies that the T-matrix obeys the following relation:

$$\text{Reciprocity:} \quad T_{-\mathbf{k}'-\sigma', -\mathbf{k}-\sigma}(-\mathbf{B}) = T_{\mathbf{k}\sigma, \mathbf{k}'\sigma'}(\mathbf{B}), \quad (2.34)$$

which is valid for *both* chiral and magneto-optical scatterers. The reciprocity principle should not be confused with time-reversal symmetry. Time reversal is stronger than reciprocity: it can be shown that the former implies the latter, but not *vice-versa* [56]. For instance, absorption (or gain) does not affect the reciprocity principle (2.34) but it destroys time reversal symmetry.

The reciprocity principle also plays a crucial role in multiple scattering and its violation has a dramatic consequence: the destruction of the coherent backscattering cone in a magnetic field. This effect was predicted theoretically [57], observed experimentally [58] and confirmed by numerical simulations [59].

Résumé du Chapitre 2 - Théorie microscopique de la diffusion simple par des particules ponctuelles

Dans ce chapitre nous allons nous intéresser à la théorie de la diffusion simple des ondes vectorielles, que nous traiterons selon une approche microscopique. Nous nous restreindrons aux *diffuseurs ponctuels*, i.e., aux diffuseurs dont la taille typique est beaucoup plus petite que la longueur d'onde de la lumière incidente. Ce genre d'approximation, qui sera utilisée tout au long de cette thèse, est extrêmement utile car elle simplifie considérablement le traitement théorique de la diffusion multiple et s'applique à un grand nombre de situations expérimentales.

Nous allons introduire le formalisme de Green et le concept de la matrice T . La matrice T est une quantité fondamentale dans la théorie de la diffusion car elle connecte les ondes incidentes et diffusées, constituant le « bloc de base » de la diffusion multiple. En outre, la matrice T contient toute l'information sur le processus de diffusion (polarisation, section efficace, etc) et reflète également les propriétés de symétrie du diffuseur (e.g., parité, réciprocity et rotation).

Le formalisme de Green et la matrice T

L'équation vectorielle de Helmholtz

$$\left[\left(\frac{\omega}{c_0} + i\epsilon \right)^2 \mathbf{I} - \nabla \times \nabla \times \right] \mathbf{G}(\omega) = \mathbf{I} + \mathbf{V}(\omega) \mathbf{G}(\omega), \quad (2.35)$$

où $\mathbf{G}(\omega)$ est le tenseur de Green (ou propagateur) et $\mathbf{V}(\omega)$, le potentiel associé à un diffuseur, peut être écrite sous la forme :

$$\mathbf{G}(\omega) = \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{G}(\omega), \quad (2.36)$$

où $\mathbf{G}_0(\omega)$ est le tenseur de Green dans le vide. L'Équation (2.36) est généralement connue comme l'équation de Lippman-Schwinger. La matrice T est définie par l'équation suivante :

$$\mathbf{G}(\omega) \equiv \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{G}_0(\omega), \quad (2.37)$$

et peut être obtenue explicitement par l'itération de $\mathbf{G}_0(\omega)$ dans l'équation intégrale (2.36), ce qui aboutit à la *série de Born* [48]:

$$\begin{aligned} \mathbf{T}(\omega) &= \mathbf{V}(\omega) + \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) + \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) + \dots \\ &\equiv t(\omega)\delta(\mathbf{r})\mathbf{I}, \end{aligned} \quad (2.38)$$

qui définit l'élément $t(\omega)$ de la matrice T . La divergence de $\mathbf{G}_0$ à $\mathbf{r} = 0$ donne une valeur nulle non-physique pour $t(\omega)$. Cela nous oblige à employer des techniques de régularisation [48] qui nous permettent d'avoir une valeur finie pour $t(\omega)$:

$$t(\omega) = -(\omega/c_0)^2 \alpha(0) \frac{\omega_0^2}{\omega_0^2 - \omega^2 - i(\Gamma\omega^3/\omega_0^2)}. \quad (2.39)$$

Au cours du processus de régularisation il est nécessaire d'introduire des paramètres pour jouer le rôle de « momentum cut-off » [48]. Ces paramètres se relient avec des quantités qui possèdent une signification physique très claire : $\alpha(0)$ et Γ , respectivement, la polarisabilité statique et la largeur de la résonance de fréquence ω_0 . Si le milieu n'est pas absorbant, $t(\omega)$ obéit au Théorème Optique [50] qui exprime la conservation d'énergie :

$$-\frac{\text{Im } t(\omega)}{k} = \frac{|t(\omega)|^2}{6\pi} = \sigma_{sca}(\omega), \quad (2.40)$$

où $\sigma_{sca}(\omega)$ est la section efficace de diffusion.

Nous pouvons finalement écrire la forme finale de la matrice T qui connecte les champs

incidents et diffusés :

$$\mathbf{T}_{tot}(\omega, r, r') = t(\omega)\delta(r - R)\delta(r' - R)\mathbf{I}. \quad (2.41)$$

Remarquons que le modèle des diffuseurs ponctuels possèdent un grand nombre d'applications, comme par exemple la diffusion de la lumière par des atomes à deux niveaux et par des petites sphères diélectriques, comme nous le verrons de façon plus détaillée au chapitre suivant.

Symétries de la matrice T

Les symétries qui gouvernent la propagation des ondes électromagnétiques se manifestent dans la matrice T de façon cruciale. Cela se voit plus facilement en écrivant la matrice T (2.41) dans la base de polarisation de van de Hulst [51]. Sur cette base, et en supposant la présence d'un champ magnétique externe $\mathbf{B}$, la matrice T a la forme :

$$T_{\mathbf{k}\sigma\mathbf{k}'\sigma'}(\mathbf{B}) = \langle \sigma(\mathbf{k}), \mathbf{k} | \mathbf{T}(\mathbf{B}, \omega) | \sigma(\mathbf{k}'), \mathbf{k}' \rangle, \quad (2.42)$$

où σ et σ' sont, respectivement, l'état d'hélicité d'onde incidente et l'état d'hélicité d'onde diffusée.

Une opération de *parité* est équivalente à une réflexion spéculaire. Si le diffuseur est symétrique sous une opération miroir, la matrice T doit donc obéir à la relation de parité :

$$\text{Parité :} \quad T_{-\mathbf{k}\sigma, -\mathbf{k}'\sigma'}(\mathbf{B}) = T_{\mathbf{k}\sigma, \mathbf{k}'\sigma'}(\mathbf{B}). \quad (2.43)$$

Un diffuseur *chiral* n'est pas invariant sous une opération miroir et par conséquent sa matrice T ne vérifie pas forcément la relation (2.43). Un diffuseur magnéto-optique, par contre, doit obéir à cette relation.

La matrice T doit également obéir au *principe de réciprocité*, qui est lié à l'échange entre la

source et le détecteur de l'onde diffusée :

$$\text{Réciprocité :} \quad T_{-\mathbf{k}'-\sigma', -\mathbf{k}-\sigma}(-\mathbf{B}) = T_{\mathbf{k}\sigma, \mathbf{k}'\sigma'}(\mathbf{B}). \quad (2.44)$$

Il est évident, à partir de la relation (2.44), que la présence d'un champ magnétique externe brise le principe de réciprocité. Ce principe ne doit pas être confondu avec la symétrie du renversement temporel [56]. Il est important de souligner que la relation (2.44) reste toujours valable pour des diffuseurs chiraux.

Finalement, il n'est pas inintéressant de noter que les relations (2.43) et (2.44) sont valables également pour des milieux absorbants, contrairement à l'Éq. (2.40).

Chapter 3

Wave transport in random media: microscopic theory of multiple scattering

After having presented a microscopic treatment of single scattering in the last chapter, we will now concentrate on wave scattering from a large number of particles. We will develop here a microscopic treatment of multiple scattering, which takes into account interference effects in wave transport. This approach is capable to describe genuine ondulatory phenomena, such as coherent backscattering and Anderson localization. The precise microscopic treatment of wave transport, based on the Green's function formalism, is extremely detailed since it requires the *a priori* knowledge of the exact positions and shapes of scatterers. This notion makes an exact microscopic approach unpractical and, in order to simplify the treatment, we will appeal to averaging techniques and controlled approximations. We will describe field propagation in disordered media by introducing the *amplitude Green's tensor averaged over disorder*, $\langle \mathbf{G} \rangle$. Subsequently, we will consider $\langle \mathbf{G} \mathbf{G}^* \rangle$ in order to describe propagation of the *averaged diffuse intensity*. The techniques present here, based on diagrammatic expansions, were originally developed in the context of (scalar) electronic transport in disordered metals [60]. The generalization of these techniques for the vector case, to describe electromagnetic wave transport, is now well very well established and can be found in standard references [49], [61].

3.1 Average amplitude: Dyson equation

The treatment of electric field (amplitude) propagation in disordered media, described by the second-rank Green tensor $\mathbf{G}(\omega)$ introduced in the last chapter, requires an average over all positions of the scatterers. The Green tensor averaged by the disorder $\langle \mathbf{G}(\omega) \rangle$ is often called the Dyson-Green tensor

$$\langle \mathbf{G}(\omega) \rangle = \left\langle \frac{1}{\varepsilon(\mathbf{x})(\omega/c_0)^2 + i\epsilon - \mathcal{A}(\mathbf{p})} \right\rangle \quad (3.1)$$

where Eq. (2.9) was used and $\varepsilon(\mathbf{x})$ and $\mathcal{A}(\mathbf{p}) = \mathbf{p}^2 - \mathbf{p}\mathbf{p}$ should be regarded as operators. By averaging Eq. (3.1) one arrives at the following expression for the Dyson-Green tensor

$$\mathbf{G}(\omega, \mathbf{p}) \equiv \frac{1}{(\omega/c_0)^2 + i\epsilon - \mathbf{p}^2 \Delta_p - \Sigma(\omega, \mathbf{p})}, \quad (3.2)$$

where the relation $\langle \mathbf{p} | \mathcal{A}(\mathbf{p}) | \mathbf{p}' \rangle = \mathbf{p}^2 \Delta_p \delta(\mathbf{p} - \mathbf{p}')$ was used. Equation (3.2) defines the *self energy* $\Sigma(\omega, \mathbf{p})$ and is called the *Dyson equation*. The self energy $\Sigma(\omega, \mathbf{p})$ represents an averaged quantity and, since averaging restores translational invariance, it can only depend on $\delta(\mathbf{p} - \mathbf{p}')$ and on frequency ω . The Dyson equation (3.2) is exact and usually it is not possible to find $\Sigma(\omega, \mathbf{p})$ and $\mathbf{G}(\omega, \mathbf{p})$ exactly. In order to solve it, one is obliged to use approximations such as the diagrammatic perturbation theory (provided the density of scatterers n is small) [60]

$$\begin{aligned} \mathbf{G}(\omega, \mathbf{p}) &= \mathbf{G}_0 + \mathbf{G}_0 \Sigma \mathbf{G}_0 + \mathbf{G}_0 \Sigma \mathbf{G}_0 \Sigma \mathbf{G}_0 + \dots \\ &= [\mathbf{G}_0^{-1}(\omega, \mathbf{p}) - \Sigma(\omega, \mathbf{p})]^{-1}, \end{aligned} \quad (3.3)$$

where $\mathbf{G}_0$ is the Green function in the vacuum (see Fig. 3-1). The self energy Σ can also be calculated via perturbation theory. In terms of diagrams, it can be represented by the so-called *irreducible diagrams* [60]. In the lowest order of the scatterer density n , Σ is proportional to the single-particle T-matrix in the forward direction $\mathbf{T}_{\mathbf{p}\mathbf{p}}(\omega)$, that was introduced in the last chapter:

$$\Sigma^1(\omega, \mathbf{p}) = n \mathbf{T}_{\mathbf{p}\mathbf{p}}(\omega). \quad (3.4)$$

$$\begin{aligned}
\text{Heavy line} &= \text{Thin line} + \text{Thin line} \times \text{Thin line} + \text{Thin line} \times \text{Thin line} \times \text{Thin line} + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} + \dots \\
&\quad + \text{Thin line} \times \text{Thin line} \times \text{Thin line} + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} \\
&\quad + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} \\
&\quad + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} \\
&\quad + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} \\
\Sigma &= \times + \text{Thin line} \times \text{Thin line} + \text{Thin line} \times \text{Thin line} \times \text{Thin line} + \text{Thin line} \times \text{Thin line} \times \text{Thin line} \times \text{Thin line} + \dots \\
&= \bullet + \bullet \times \bullet \times \bullet + \dots \\
\text{Heavy line} &= \text{Thin line} + \text{Thin line} \times \Sigma + \text{Thin line} \times \Sigma \times \Sigma + \text{Thin line} \times \Sigma \times \Sigma \times \Sigma + \dots
\end{aligned}$$

Figure 3-1: Representation of amplitude diagrams: the averaged Green's tensor $\langle \mathbf{G}(\omega, \mathbf{p}) \rangle$ (heavy line) in disordered media. The thin lines represent the Green's tensor in free space $\mathbf{G}_0(\omega, \mathbf{p})$ (also called bare Green's tensors); the crosses represent the potential V and the dashed lines indicate that the wave is scattered by the same particle. Below, the self-energy Σ is depicted as a sum of irreducible diagrams; the full circles represent the T matrices. In the Independent Scattering Approximation one neglects recurrent scattering (represented by the last diagram in the right) and Σ turns out to be proportional to the T matrix in the forward direction. The lowest line represents the average Green's tensor $\langle \mathbf{G}(\omega, \mathbf{p}) \rangle$ constructed from the self-energy Σ .

This low-density approximation is called the *independent scatterer approximation* (ISA). Equation (3.4) clearly shows that single scattering (associated with $\mathbf{T}_{\mathbf{p}\mathbf{p}}(\omega)$) constitutes the “building block” for multiple scattering (associated with $\Sigma^1(\omega, \mathbf{p})$). The ISA takes into account all orders of multiple scattering, but ignores recurrent scattering events, i.e., events in which the wave is scattered more than once by the same particle. For several applications in multiple light scattering, recurrent scattering can be disregarded and the ISA applies. However, for very strong scattering media, specially near to the transition to Anderson localization, recurrent scattering becomes important. Manifestations of recurrent scattering and the consequent breakdown of the ISA in enhanced backscattering cone have been experimentally observed [62].

In view of the average in Eq. (3.3), the scattering medium can be regarded as an effective

medium with dielectric constant defined by

$$\varepsilon(\omega, \mathbf{p}) = \mathbf{I} - \frac{\Sigma(\omega, \mathbf{p})}{(\omega/c_0)^2}. \quad (3.5)$$

One should realize that averaging is a dissipative process and consequently it involves a loss of information. As a result, the dielectric tensor (3.5) has a *positive* imaginary part, which can be regarded as a manifestation of the dissipation-fluctuation theorem [13]. Physically, the poles of the Dyson Green tensor (3.3) determine the excitations of light propagation that, due to the positive imaginary part of Eq. (3.5), will decay in space. Decay and propagation of electromagnetic modes are characterized by the complex index of refraction

$$m^2(\omega) \equiv \varepsilon(\omega) = K^2(\omega)(c_0/\omega)^2, \quad (3.6)$$

where K is the effective wave number. It defines the phase velocity v_p and the extinction mean free path ℓ according to:

$$K(\omega) = \frac{\omega}{v_p(\omega)} + \frac{i}{2\ell(\omega)}. \quad (3.7)$$

The averaged Green's tensor in the coordinate space (and in the far field regime $kr \gg 1$) is given by [49]

$$\langle \mathbf{G}(\omega, \mathbf{r}) \rangle = -\frac{\Delta_{\mathbf{r}}}{4\pi r} \exp(i\omega r/v_p) \exp(-r/2\ell). \quad (3.8)$$

Physically, Eq. (3.8) describes the decay of the coherent field amplitude $\mathbf{E}(\omega, \mathbf{r})$. The length scale that characterizes this decay is the extinction mean free path ℓ , as expressed by Eq. (3.8). It should be stressed here that this decay (extinction) is not necessarily due to absorption, but also due to scattering out of the forward direction.

3.2 Average intensity: Bethe-Salpeter equation

Having formulated a microscopic description for the transport of the averaged amplitude, we can now focus on the description of the averaged intensity. Both these two quantities are

fundamental in transport theory and both are experimentally accessible for light. The treatment here will be quite similar to the one applied for the averaged amplitude discussed in the last section.

The quantity of interest here is ensemble averaged intensity $\langle I(\mathbf{r}, t) \rangle$ which is proportional to the absolute square of the electric field amplitude $\mathbf{E}(\mathbf{r}, t)$

$$\langle I(\mathbf{r}, t) \rangle = \left\langle |\mathbf{E}(\mathbf{r}, t)|^2 \right\rangle. \quad (3.9)$$

This quantity involves the product $\langle \mathbf{G}(\mathbf{r}, \mathbf{r}') \mathbf{G}^*(\mathbf{r}, \mathbf{r}') \rangle$ of the Green tensor and its complex conjugate, which is associated with the propagation of the field. By performing a Fourier-Laplace transform in the temporal part of Eq. (3.9) and expressing its spatial part in the momentum representation, one obtains [49]:

$$\begin{aligned} & \langle \mathbf{G}(\omega + \Omega/2 + i\epsilon, \mathbf{p}_+, \mathbf{p}'_+) \mathbf{G}^*(\omega - \Omega/2 - i\epsilon, \mathbf{p}_-, \mathbf{p}'_-) \rangle \\ & \equiv \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) \times (2\pi)^3 \delta(-\mathbf{p}_+ + \mathbf{p}'_+ + \mathbf{p}_- - \mathbf{p}'_-). \end{aligned} \quad (3.10)$$

The momentum variables $\mathbf{p}$, $\mathbf{p}'$ and $\mathbf{q}$ are defined according to $\mathbf{p}_\pm = \mathbf{p} \pm \mathbf{q}/2$ and $\mathbf{p}'_\pm = \mathbf{p}' \pm \mathbf{q}/2$. Physically, Eq. (3.10) describes a wave packet characterized by internal oscillations with frequency ω (in time) and wave vector $\mathbf{p}$ (in space). The frequency Ω and the wave vector $\mathbf{q}$ are associated with the oscillations in time and space of the *envelope* of the wave packet, respectively. Typically, the variables Ω and $\mathbf{q}$ denote oscillations that vary much more slowly than the cycles described by internal variables ω and $\mathbf{p}$. This gives rise to the “Slowly Varying Envelope Approximation” (SVEA). Since the momentum $\mathbf{p}'$ describes the spatial characteristics of the source, we will sum it to get the response of a point-source:

$$\Phi_{\omega \mathbf{p}}(\Omega, \mathbf{q}) = \sum_{\mathbf{p}'} \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) \quad (3.11)$$

Equation (3.11) represents the exact microscopic definition of the specific intensity $I_{\omega \mathbf{p}}(\mathbf{q})$ appearing in radiative transport theory and constitutes the fundamental transport quantity [49]. We introduce two important physical quantities that will be useful in the following: the average electromagnetic energy density $\rho_\omega(\Omega, \mathbf{q})$ and the current density $\mathbf{J}_\omega(\Omega, \mathbf{q})$ at frequency

ω . They are defined according to:

$$\rho_{\omega}(\Omega, \mathbf{q}) \equiv (\omega/c_0)^2 \sum_{\mathbf{p}} \Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q}), \quad (3.12)$$

$$\mathbf{J}_{\omega}(\Omega, \mathbf{q}) = \omega \sum_{\mathbf{p}} \mathbf{p} \Phi_{\omega\mathbf{p}}(\Omega, \mathbf{q}). \quad (3.13)$$

We will also introduce a new object to describe the transport of the averaged intensity, the *Irreducible Vertex* $\mathbf{U}$, which plays the same role of the self energy Σ for the averaged amplitude. It is defined by the equation

$$\langle \mathbf{G}(\omega) \mathbf{G}^*(\omega') \rangle = \langle \mathbf{G}(\omega) \rangle \langle \mathbf{G}^*(\omega') \rangle + \langle \mathbf{G}(\omega) \rangle \langle \mathbf{G}^*(\omega') \rangle \cdot \mathbf{U}(\omega, \omega') \cdot \langle \mathbf{G}(\omega) \mathbf{G}^*(\omega') \rangle, \quad (3.14)$$

called the *Bethe-Salpeter equation*. It represents the analogy of the Dyson equation (3.3) for the averaged amplitude: just as the amplitude obeys the Dyson equation, the intensity obeys the Bethe-Salpeter equation. Like the self energy Σ , the four-rank vertex $\mathbf{U}$ can be represented diagrammatically by irreducible diagrams.

The diagrammatic representation of the averaged intensity is depicted in Fig. 3-2. In order to calculate the intensity, we must multiply the Green's tensor $\mathbf{G}$ by its complex conjugate $\mathbf{G}^*$. Diagrammatically, this is represented by drawing the expansion for $\mathbf{G}$ above and drawing the one for $\mathbf{G}^*$ below, as represented by solid lines in Fig. 3-2. The scattering events are represented by full circles. Dashed lines connecting upper and bottom solid lines denote the situation where $\mathbf{G}$ and $\mathbf{G}^*$ scatterer from a common scatterer. One can identify two fundamental types of intensity diagrams: the ladder and the most crossed diagrams. The ladder diagrams describe the averaged diffuse intensity in the ISA: they correspond to the scenario of intensity propagation from one scatterer to another. According to Fig. 3-2, the intensity can be decomposed into terms without scattering, terms with one common scattering event, terms with two common scattering events and so on. This situation gives rise to the following integral equation (in momentum space) for the ladder four-rank tensor $\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q})$ [61]:

$$\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q}) = n \mathbf{T}_{\mathbf{p}\mathbf{p}'} \otimes \mathbf{T}_{\mathbf{p}\mathbf{p}'}^* + n \sum_{\mathbf{p}''} \mathbf{T}_{\mathbf{p}\mathbf{p}''} \cdot (\mathbf{G}(\mathbf{p}''_+) \otimes \mathbf{G}^*(\mathbf{p}''_-)) \cdot \mathbf{T}_{\mathbf{p}\mathbf{p}''}^* \cdot \mathcal{L}(\mathbf{p}'', \mathbf{p}', \mathbf{q}). \quad (3.15)$$

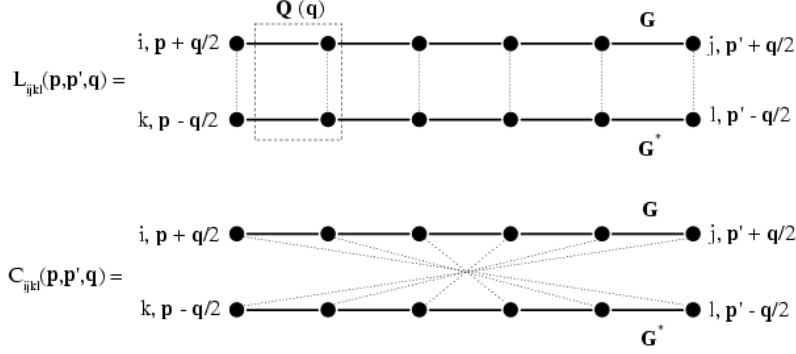


Figure 3-2: Representation of two important intensity diagrams: ensemble averaged ladder $\mathbf{L}$ (top) and most crossed $\mathbf{C}$ tensors (bottom). The averaged intensity is calculated by multiplying the Green's tensor $\mathbf{G}$ by its complex conjugate $\mathbf{G}^*$, represented diagrammatically by upper and lower solid line in each intensity diagram, respectively. Full circles represent T matrices. The indexes i, k (j, l) describe the state of polarization of the incident (emergent) radiation. The tensor $\mathbf{Q}$ constitutes the “building block” of ladder diagrams.

The so-called most crossed diagrams (also exhibited in Fig. 3-2), not contained into the set of ladder diagrams, are responsible for enhanced backscattering and can be obtained from the ladder diagrams by a reciprocity operation.

3.2.1 Boltzmann approximation

Like any exact microscopic equation, Eq. (3.27) requires the employment of approximations in order to be solved. In the lowest order of the scatterers density n , the Irreducible Vertex takes the form

$$\mathbf{U}_{\omega, \mathbf{p}\mathbf{p}'}^1(\Omega, \mathbf{q}) = n \mathbf{T}_{\mathbf{p}_+ \mathbf{p}'_+} \mathbf{T}_{\mathbf{p}_- \mathbf{p}'_-}^* + \mathcal{O}(n^2), \quad (3.16)$$

which constitutes the *Boltzmann Approximation*. Using this approximation, the Bethe-Salpeter equation can be reduced to the Equation of Radiative Transfer for the specific intensity [49]. Physically, the Boltzmann approximation corresponds, at the level of intensity transport, to the assumption that each scatterer is visited only once, and that no interference takes place.

3.2.2 Diffusion approximation

Even after having applied the Boltzmann approximation, it is very hard to obtain analytical solutions for the Bethe-Salpeter equation and numerical methods should be employed. In order to derive analytical solutions even for common geometries and simplified T-matrices, one should rely on a further simplification: the *diffusion approximation*. This approximation is valid for small Ω (much smaller than ω) and for small $\mathbf{q}$ (much smaller than $\mathbf{p}$). It works quite well experimentally [63], although some situations have been reported where it is believed to break down do exist [64].

In a diagrammatic treatment of the averaged intensity, the diffusion approximation consists in expanding Eq. (3.15) for small $\mathbf{q}$ (the so-called hydrodynamic limit), keeping up to quadratic terms in $\mathbf{q}$ and summing the resulting series. For simplicity, we will consider the case of point scatterers for which the T-matrix t has a simple form, independent of $\mathbf{p}$ and $\mathbf{p}'$. Introducing the four-rank tensor $\mathbf{Q}(\mathbf{q})$ that represents “building block” of ladder diagrams (see Fig. 3-2) and expanding it in powers of $\mathbf{q}$ one obtains (up to order $\mathbf{q}^2$) [57]

$$Q(\mathbf{q})_{ijkl} \sim n |t|^2 \int G_{ij}(\mathbf{p} + \mathbf{q}/2) G_{lk}^*(\mathbf{p} - \mathbf{q}/2) d^3\mathbf{p} \simeq \delta_{ij} \delta_{kl} (1 - \mathbf{q}^2 \ell^2) + \dots \quad (3.17)$$

In terms of $\mathbf{Q}(\mathbf{q})$, it turns out that the ladder tensor $\mathcal{L}$ has the form of a geometric series with ratio $\mathbf{Q}(\mathbf{q})$

$$\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q}) \sim \frac{1}{\ell(1 - \mathbf{Q}(\mathbf{q}))} = \frac{3}{\ell^3 \mathbf{q}^2}. \quad (3.18)$$

This equation expresses that, within the diffusion approximation, the ladder tensor $\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q})$ (3.18) has a singularity at $\mathbf{q} = \mathbf{0}$. This singularity is associated with long-range diffusion in infinite media but disappears in finite media.

It is possible to show, using rigorous transport theory, that the ladder tensor $\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q})$ can

be expressed as [65]

$$\mathcal{L}_{ijkl}(\mathbf{p}, \mathbf{p}', \mathbf{q}) = \frac{2\pi}{\ell^2} \frac{d_{ik}(\mathbf{p}, \mathbf{q}) d_{lj}(-\mathbf{p}', -\mathbf{q})}{D(\mathbf{q})}, \quad (3.19)$$

which is equivalent to the diffusion equation. In Eq. (3.19), the second rank tensor $\mathbf{d}(\mathbf{p}, \mathbf{q})$ is the eigenvector of the ladder tensor associated with eigenvalue of $\mathbf{Q}(\mathbf{q})$ close to 1. The tensor $\mathbf{d}(\mathbf{p}, \mathbf{q})$ describes the specific intensity at the direction $\mathbf{p}$. $D(\mathbf{q})$ is some scalar function that is *at least* linear in $\mathbf{q}$ if no absorption occurs. In the absence of chirality (see Chapter 5), symmetry arguments impose that $D(\mathbf{q})$ must have the leading bilinear form $D(\mathbf{q}) = \mathbf{q} \cdot \mathbf{D} \cdot \mathbf{q}$, with $\mathbf{D}$ the diffusion tensor. Actually, the form of the numerator of Eq. (3.19) is imposed by symmetry relations, namely parity and reciprocity, that $\mathcal{L}(\mathbf{p}, \mathbf{p}', \mathbf{q})$ must obey [56]

$$\begin{aligned} \text{Reciprocity} \quad : \quad & \mathcal{L}_{ijkl}(\mathbf{p}, \mathbf{p}', \mathbf{q}) = \mathcal{L}_{jilk}(-\mathbf{p}', -\mathbf{p}, -\mathbf{q}) \\ \text{Parity} \quad : \quad & \mathcal{L}_{ijkl}(\mathbf{p}, \mathbf{p}', \mathbf{q}) = \mathcal{L}_{ijkl}(-\mathbf{p}, -\mathbf{p}', -\mathbf{q}) \end{aligned} \quad (3.20)$$

The symmetry relations (3.20) are equally valid for the Irreducible Vertex $\mathbf{U}$ and express the symmetry properties of the T-matrix (see Eqs. (2.33) and (2.34)).

Résumé du Chapitre 3 - Transport des ondes dans les milieux inhomogènes : théorie microscopique de la diffusion multiple

Après avoir présenté la théorie microscopique de la diffusion simple au chapitre 2, nous allons nous centrer, ici, sur la théorie microscopique de la diffusion multiple. Cette théorie prend en compte les effets d'interférence du transport de la lumière et est donc capable de décrire les phénomènes physiques manifestement ondulatoires, comme la rétrodiffusion cohérente [66][67] et la localisation forte d'Anderson [10]. La théorie microscopique exacte du transport de lumière dans les milieux désordonnés est relativement compliquée car elle requiert *a priori* la connaissance de toutes les positions des diffuseurs ainsi que la forme précise de ses matrices T . A cause de ce type de contraintes, ce genre de traitement reste peu pratique en termes d'application et l'emploi de techniques de moyennement et d'approximations s'impose. Nous allons donc introduire, dans un premier temps, le tenseur d'amplitude de Green moyenné sur le désordre, $\langle \mathbf{G} \rangle$, puis nous centrer sur la description de l'intensité lumineuse moyenne en utilisant la quantité $\langle \mathbf{G} \mathbf{G}^* \rangle$. Soulignons que le formalisme présenté ici, basé sur les expansions diagrammatiques, a été développé originalement dans le cadre du transport (scalaire) électronique dans des métaux désordonnés [60]. La généralisation de ce formalisme au cas vectoriel pour décrire le transport des ondes électromagnétiques est aujourd'hui très bien établie et peut être trouvée dans des références standards [49][61]. Nous ne ferons ici qu'une brève description de la théorie

microscopique du transport d'ondes électromagnétiques.

Amplitude moyenne : l'équation de Dyson

Le traitement de la propagation de l'amplitude moyenne du champ électrique dans un milieu désordonné se fait à partir du tenseur de Green $\mathbf{G}(\omega)$ moyenné sur le désordre (également appelé tenseur de Dyson-Green) :

$$\begin{aligned}\langle \mathbf{G}(\omega) \rangle &= \left\langle \frac{1}{\varepsilon(\mathbf{x})(\omega/c_0)^2 + i\epsilon - \mathbf{p}^2 + \mathbf{p}\mathbf{p}} \right\rangle \\ &\equiv \frac{1}{(\omega/c_0)^2 + i\epsilon - \mathbf{p}^2 \Delta_p - \Sigma(\omega, \mathbf{p})}.\end{aligned}\quad (3.21)$$

L'équation (3.21) s'appelle l'équation de Dyson et définit la self-énergie $\Sigma(\omega, \mathbf{p})$. À la condition que la densité des diffuseurs soit faible, nous pouvons faire appel à la théorie de perturbations diagrammatiques [60] pour résoudre l'éq (3.21) :

$$\begin{aligned}\langle \mathbf{G}(\omega, \mathbf{p}) \rangle &= \mathbf{G}_0 + \mathbf{G}_0 \Sigma \mathbf{G}_0 + \mathbf{G}_0 \Sigma \mathbf{G}_0 \Sigma \mathbf{G}_0 + \dots \\ &= [\mathbf{G}_0^{-1}(\omega, \mathbf{p}) - \Sigma(\omega, \mathbf{p})]^{-1},\end{aligned}\quad (3.22)$$

où $\mathbf{G}_0$ est la fonction de Green dans le vide.

La self-énergie Σ peut, elle aussi, être évaluée à l'aide de la théorie des perturbations et représente, en termes de diagrammes, la somme de tous les diagrammes irréductibles (i.e., que l'on ne peut pas découper en diagrammes plus simples). À l'ordre le plus bas de la densité n , la self-énergie est proportionnelle à la matrice T vers l'avant $\mathbf{T}_{\mathbf{p}\mathbf{p}}(\omega)$:

$$\Sigma^1(\omega, \mathbf{p}) = n \mathbf{T}_{\mathbf{p}\mathbf{p}}(\omega). \quad (3.23)$$

L'approximation (3.23) s'appelle « *Approximation de Diffusion Indépendante* » et signifie que la diffusion a lieu de façon indépendante sur chaque diffuseur (i.e., chaque diffuseur n'est visité qu'une seule fois).

Après la moyenne en Eq. (3.22), le milieu désordonné peut être vu comme un milieu effectif : les pôles du tenseur de Dyson-Green $\langle \mathbf{G}(\omega, \mathbf{r}, \mathbf{p}) \rangle$ déterminent les excitations de la propagation

de la lumière. Il est important de remarquer que la moyenne est un processus dissipatif et, par conséquent, les excitations du milieu iront forcément décroissant dans l'espace. En effet, le tenseur de Green moyen dans l'espace réel (et dans le régime du champ lointain $kr \gg 1$) s'écrit comme :

$$\langle \mathbf{G}(\omega, \mathbf{r}) \rangle = -\frac{\Delta_{\mathbf{r}}}{4\pi r} \exp(i\omega r/v_p) \exp(-r/2\ell). \quad (3.24)$$

L'équation (3.24) décrit la décroissance du faisceau cohérent avec la longueur caractéristique ℓ , le libre parcours moyen.

Intensité moyenne : l'équation de Bethe-Salpeter

Le traitement de l'intensité moyenne requiert la multiplication du tenseur de Green $\mathbf{G}(\mathbf{r}, \mathbf{r}')$ par son complexe conjugué $\mathbf{G}^*(\mathbf{r}, \mathbf{r}')$, $\langle \mathbf{G}(\mathbf{r}, \mathbf{r}') \mathbf{G}^*(\mathbf{r}, \mathbf{r}') \rangle$. En réalisant une transformation Laplace-Fourier en $\langle \mathbf{G}(\mathbf{r}, \mathbf{r}') \mathbf{G}^*(\mathbf{r}, \mathbf{r}') \rangle$ et en exprimant la partie spatiale dans la représentation des momenta, on arrive à [49] :

$$\begin{aligned} & \langle \mathbf{G}(\omega + \Omega/2 + i\epsilon, \mathbf{p}_+, \mathbf{p}'_+) \mathbf{G}^*(\omega - \Omega/2 - i\epsilon, \mathbf{p}_-, \mathbf{p}'_-) \rangle \\ & \equiv \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}) \times (2\pi)^3 \delta(-\mathbf{p}_+ + \mathbf{p}'_+ + \mathbf{p}_- - \mathbf{p}'_-), \end{aligned} \quad (3.25)$$

où les variables $\mathbf{p}$, $\mathbf{p}'$ et $\mathbf{q}$ sont définies comme $\mathbf{p}_{\pm} = \mathbf{p} \pm \mathbf{q}/2$ et $\mathbf{p}'_{\pm} = \mathbf{p}' \pm \mathbf{q}/2$. Physiquement, Eq. (3.25) représente un paquet d'onde avec des oscillations internes de fréquence ω (en temps) et de vecteur d'onde $\mathbf{p}$ (en espace). En sommant sur $\mathbf{p}'$, qui décrit les caractéristiques de la source, on obtient :

$$\Phi_{\omega \mathbf{p}}(\Omega, \mathbf{q}) = \sum_{\mathbf{p}'} \Phi_{\omega, \mathbf{p}\mathbf{p}'}(\Omega, \mathbf{q}). \quad (3.26)$$

La quantité $\Phi_{\omega \mathbf{p}}(\Omega, \mathbf{q})$ en (3.26) constitue la quantité fondamentale de la théorie microscopique du transport de la lumière et représente (à quelques constantes près) la définition microscopique exacte de l'intensité spécifique qui figure dans la théorie du transfert radiatif [68].

Nous pouvons maintenant procéder de façon similaire au cas de l'amplitude moyenne présen-

tée antérieurement et définir l'équivalent de la self-énergie Σ pour l'intensité moyenne, le *Vertex Irréductible* $\mathbf{U}$:

$$\langle \mathbf{G}(\omega) \mathbf{G}^*(\omega') \rangle = \langle \mathbf{G}(\omega) \rangle \langle \mathbf{G}^*(\omega') \rangle + \langle \mathbf{G}(\omega) \rangle \langle \mathbf{G}^*(\omega') \rangle \cdot \mathbf{U}(\omega, \omega') \cdot \langle \mathbf{G}(\omega) \mathbf{G}^*(\omega') \rangle. \quad (3.27)$$

Équation (3.27) s'appelle *équation de Bethe-Salpeter*. Ainsi comme la self-énergie Σ , le *Vertex Irréductible* $\mathbf{U}$ ne contient que des diagrammes irréductibles.

Chapter 4

Light scattering in magnetochiral random media

Magnetochiral scattering of light: Optical manifestation of chirality

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Abstract

We have investigated multiple scattering of light in systems subject to magnetochiral (MC) effects. Our medium consists of magneto optically active dipoles placed in a chiral geometry under the influence of an external magnetic field. We have calculated the total and the differential scattering MC cross sections of this system, explicitly showing that they are proportional to *pseudoscalar* quantities. This provides a new optical measure for the degree of chirality, a pseudoscalar g , of an arbitrary geometrical configuration of scatterers based on its scattering properties. We have calculated g for some simple chiral systems and we have even used it to probe the degree of optical chirality of random systems. Finally, we have compared g with other recently defined chiral measures in literature.

Résumé du Chapitre 4 - Diffusion de la lumière dans les milieux magnéto-chiraux : une nouvelle manifestation de la chiralité en optique

Ce chapitre sera consacré aux études de la diffusion multiple de la lumière dans les milieux magnéto-chiraux. À partir des caractéristiques de l'effet magnéto-chiral dans les milieux homogènes, décrites au Chapitre 1, nous développerons un modèle pour étudier cet effet dans les *milieux inhomogènes*. Dans le cadre de ce modèle, nous allons montrer que les quantités qui caractérisent le processus de diffusion, comme par exemple les sections efficaces, sont proportionnelles aux grandeurs *pseudo-scalaires* et par conséquent constituent une nouvelle manifestation de la chiralité en optique, comme l'activité optique naturelle. Nous allons exploiter ce fait pour la construction d'une nouvelle mesure du degré de chiralité associé à une configuration arbitraire de diffuseurs : le pseudo-scalaire g . En utilisant le paramètre g , on s'attachera à étudier le degré de chiralité de quelques systèmes simples et même de systèmes désordonnés. On a également comparé g avec les mesures de la chiralité introduites récemment par d'autres auteurs.

Description du modèle

Afin de générer des effets magnéto-chiraux en diffusion multiple, remarquons d’abord que cet effet repose, comme nous l’avons remarqué au chapitre 1, sur des conditions de symétrie très particulières : il ne se manifeste que pour les matériaux où les symétries miroir et du renversement du temps sont simultanément brisées. Nous proposons un scénario simple pour modéliser l’effet magnéto-chiral dans le cadre de la diffusion multiple de la lumière : diffuseurs magnéto-optiques placés dans des configurations géométriques chirales. Avant de décrire quantitativement le modèle proposé, il est instructif de faire le point sur le concept de la chiralité et sur quelques tentatives récentes de la quantifier.

Le concept de la chiralité et les paramètres chiraux

La plupart des objets naturels ne sont pas superposables à leur image dans un miroir, comme par exemple nos mains ou les vis. Ils sont dits *chiraux* (du grec *kheir*, main) et se présentent sous formes « gauche » ou « droite », fréquemment appelées formes enantiomères (du grec *enantios*, opposé). L’expression *chirale* a été créée par Lord Kelvin en 1893:

« I call any geometrical figure, or group of points, chiral, and say it has chirality if its image in a plane mirror, ideally realized, cannot be brought to coincide with itself. »

La notion de chiralité est fondamentale et ubiquie dans la nature, avec des profondes implications dans divers domaines de la recherche : la recherche pharmaceutique, la physique de la matière molle [69] et dans les sciences du vivant. Dans le dernier cas, en particulier, notons que plusieurs types de coquillages, quelques types de plantes, bactéries et animaux présentent une forme géométrique chirale. L’homme aussi est anatomiquement chiral : son coeur est à gauche et son foie est à droite. Pasteur fut le premier à mettre en évidence le rôle déterminant de l’asymétrie dans les systèmes vivants. Il a constaté que l’acide tartarique est produit par voie physico-chimique sous deux formes cristallines chirales enantiomères. Pourtant, il a également observé, à partir d’expériences sur le pouvoir rotatoire, que cette même substance ne se produit que dans l’une des deux formes chirales par la voie biologique. Cela lui a permis d’inférer que

l'asymétrie des molécules organiques du monde vivant est le concept qui différencie le mieux la matière vivante de la matière inerte. Cette conclusion a été corroborée plus tard avec la découverte que les êtres vivants n'utilisent que les aminoacides « gauches » pour synthétiser les protéines et que l'homochiralité est nécessaire pour produire la forme précise des enzymes et de l'ADN. L'origine de la « homochiralité de la vie » constitue encore une des grandes questions ouvertes de la science pour laquelle l'effet magnéto-chiral peut offrir une réponse, comme cela a été discuté au paragraphe 1.3.2. Pour une discussion plus complète et plus générale du concept de la chiralité dans la nature, on renvoie le lecteur aux refs. [70] et [71].

Malgré l'importance de la chiralité en science, il peut paraître étonnant que récemment seulement il y ait eu des progrès dans le sens de la « quantification » ou de la « mesure » du degré de chiralité associé à un objet quelconque, c'est-à-dire dans la définition d'un « paramètre d'ordre » pour la chiralité, comme on peut définir le degré d'ordre pour un matériel ferromagnétique. L'une des premières tentatives d'établir une mesure quantitative de la chiralité revient à Gilat [72]. Osipov *et al.* [73] et plus récemment Solymosi *et al.* [74] ont proposé des paramètres chiraux qui reposaient sur des modèles simples de l'activité optique naturelle. Harris *et al.* [75][76][77] ont introduit une approche élégante, basée sur la théorie des groupes, pour quantifier la chiralité associée à un objet ou un ensemble de points. Ils ont construit des quantités pseudo-scalaires invariantes sur rotations pour déterminer la chiralité de certaines molécules et ils ont montré comment ces quantités interviennent dans quelques observables chirales, comme par exemple le « pitch » d'un cristal liquide cholestérique [75] et le pouvoir rotatoire optique [78]. Récemment, le problème de la quantification de la chiralité a également attiré l'attention des mathématiciens [79][80].

Il existe bien sûr plusieurs possibilités pour définir un paramètre d'ordre chiral, le choix n'étant pas unique. Néanmoins, un paramètre chiral doit forcément être une quantité pseudo-scalaire, invariante sous rotations. Avec cette dernière contrainte, on garantit qu'il n'existe aucune rotation capable de faire coïncider l'image miroir d'un objet chiral à lui-même, ce qui est consistant avec la définition de Lord Kelvin.

Par la suite, nous nous servirons de ces concepts généraux sur la chiralité pour construire une nouvelle mesure pour le degré de chiralité, basée sur l'effet magnéto-chiral.

Diffusion multiple de la lumiere dans les milieux magnéto-chiraux

Afin d'établir une mesure quantitative du degré de chiralité en exploitant notre modèle pour l'effet magnéto-chiral dans les milieux inhomogènes, il faut d'abord considérer comment un ensemble de particules magnéto-optiques sous champ magnétique $\mathbf{B}$ diffusent la lumière. Nous allons considérer des *diffuseurs ponctuels*, i.e., des particules dont la taille typique est beaucoup plus petite que la longueur d'onde incidente λ . La matrice T (3×3) d'un tel diffuseur, jusqu'au premier ordre en $\mathbf{B}$, s'écrit ainsi [54] :

$$\mathbf{t}(\mathbf{B}, \omega) = t_0 \mathbf{U} + t_1 \Phi, \quad (4.1)$$

où $\mathbf{U}$ est la matrice d'unité et le tenseur Φ a été défini en (1.12). Le paramètre t_0 n'est rien d'autre que la matrice T d'un diffuseur Rayleigh ordinaire [54]. La quantité t_1 est définie comme $t_1 = -\alpha(B)kt_0^2/4\pi$ où $\alpha(B)$ est un parametre magnéto-optique. Pour un atome, $\alpha(B)$ est associée au Zeeman splitting $zB = \Delta E_z/\hbar$ (typiquement $\alpha = zB/\Gamma$, où Γ est l'élargissement de la résonance atomique).

La connaissance de la matrice T pour la diffusion simple (4.1) nous permet d'écrire la matrice T ($3N \times 3N$) totale pour un système de N diffuseurs :

$$\mathbf{M}(k) \equiv \mathbf{t}(k) \cdot [\mathbf{U} - \mathbf{G}(k) \cdot \mathbf{t}(k)]^{-1}, \quad (4.2)$$

où $\mathbf{G}(k)$ est le tenseur de Green[49]. La diagonalisation de la matrice $\mathbf{M}(k)$ en (4.2) fournit l'amplitude et la phase du champ électrique diffusé par l'ensemble des particules [81][82]. A partir de $\mathbf{M}(k)$ il est possible de déterminer la *section efficace totale de diffusion* $\sigma_{ext}(\mathbf{B}, \mathbf{k})$ via le théorème optique (voir chapitre 2 et Ref. [50]). Comme nous nous intéressons aux effets magnéto-chiraux, il est nécessaire de calculer la partie *linéaire en $\mathbf{B}$* de la section efficace totale, $\sigma_{ext}^1(\mathbf{B}, \mathbf{k})$. En outre, comme nous voulons exploiter les propriétés de diffusion pour définir une grandeur sensible au degré de chiralité, il faut moyennner σ_{ext}^1 sur *toutes les rotations solides*. Cela revient à calculer $\langle \sigma_{ext}^1(\mathbf{B}, \mathbf{k}) \rangle_{\mathbf{k} \in 4\pi}$ qui s'écrit ainsi :

$$\langle \sigma_{ext}^1(\mathbf{B}, \mathbf{k}) \rangle_{4\pi} = g\alpha(B)(\hat{\mathbf{B}} \cdot \hat{\mathbf{k}}) \quad (4.3)$$

où g est un *pseudo-scalaire*. Signalons que l'Eq. (4.3) est manifestement une quantité invariante sur rotations et obéit aux deux relations fondamentales de symétrie des milieux magnéto-chiraux (la parité et la reciprocité), comme il est facile de vérifier. Le pseudo-scalaire g peut être exprimé en fonction de la matrice $\mathbf{M}(k)$ et des positions des diffuseurs.

Pour sonder la chiralité via l'effet magnéto-chiral

Le but de ce paragraphe est d'appliquer les résultats précédents, notamment l'Eq. (4.3), et de montrer qu'ils constituent effectivement une mesure adéquate du degré de chiralité des systèmes diffuseurs. Nous envisagerons aussi de comparer nos résultats avec les paramètres chiraux qui ont été proposés récemment par d'autres auteurs.

Nous avons calculé numériquement le pseudo-scalaire g pour des systèmes chiraux simples, comme le « twisted H », et aussi pour des systèmes désordonnés qui possèdent, en général, un caractère chiral. Il est évident que g doit s'annuler pour des systèmes diffuseurs composés de $N = 2, 3$ particules car il est impossible de générer une configuration chirale avec seulement deux ou trois particules. En effet, avec une simple rotation on peut toujours faire coïncider l'image miroir d'un système de deux particules avec lui-même. En outre, un système composé de trois particules est forcément achiral car il est toujours contenu dans un plan qui est automatiquement son plan miroir. Le système chiral le plus simple doit donc être composé d'au moins quatre particules, comme il est le cas pour le « twisted H ». Pour quatre diffuseurs magnéto-optiques distribués dans la géométrie d'un « twisted H », nous avons montré que g varie, pour certaines valeurs de la longueur d'onde incidente, comme $g \sim \sin(\gamma)$ où γ est l'angle entre les bras du « twisted H ». Remarquons que Harris *et al.* [75][76][77] ont défini un paramètre chiral ψ que ne dépend que des positions des particules et qui a pour le « twisted H » la même dépendance synodale en γ . Cela nous pousse à examiner de façon plus détaillée les connexions possibles qui existent entre ψ et g pour des géométries plus complexes.

Pour cela, nous avons évalué les paramètres g et ψ pour des systèmes de N particules aléatoirement distribuées dans une sphère de rayon R en utilisant une simulation de Monte Carlo. Nous avons pris en compte 1000 réalisations différentes du désordre, ce qui nous a permis de calculer les variances Δg et $\Delta \psi$ des paramètres chiraux g et ψ . Nous avons conclu que g et ψ sont *statistiquement proportionnels*. La relation de proportionnalité observée est la

suivante :

$$\frac{\Delta g}{\sigma_0} \propto \frac{\Delta \psi}{R^8} \lambda^3. \quad (4.4)$$

La relation ci-dessus montre qu'il est possible d'établir une correspondance entre le paramètre chiral optique g et le paramètre chiral purement géométrique ψ/R^8 si nous multiplions le dernier par la volume optique λ^3 . Finalement, ce résultat montre aussi que la correspondance entre g et ψ n'est pas seulement valable pour le cas particulier du « twisted H », mais s'applique même aux systèmes désordonnés.

Chapter 5

Light propagation in chiral and magnetochiral random media

Light transport in chiral and magnetochiral random media

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Abstract

We present a microscopic approach to study electromagnetic wave propagation in media with broken mirror symmetry. We introduce and calculate the transport mean free path ℓ_C^* associated with the residual polarization of diffuse light in chiral systems. In chiral media subject to an external magnetic field $\mathbf{B}$, all symmetry requirements exist to create a macroscopic "super" light current in the direction of $\mathbf{B}$ that persists even in the absence of a spatial photon density gradient. However, we show that such a current is identically zero in our model. We finally show the existence of a linear magneto transmission in magnetochiral media.

Résumé du Chapitre 5 - Transport de la lumière dans les milieux chiraux et magnéto-chiraux

Le but de ce chapitre est d'établir une description microscopique de la diffusion multiple de la lumière dans les milieux chiraux et magnéto-chiraux. La théorie vectorielle de la diffusion multiple de la lumière, dont les principaux concepts ont été présentés au chapitre 3, est aujourd'hui très bien établie [61]. La base microscopique de cette théorie repose sur l'équation de Bethe-Salpeter (EBS), développée originalement dans le cadre de la théorie de la conduction électronique [60], et qui a été appliquée pour décrire la propagation des ondes scalaires [83][84][85], électromagnétiques [65], et même en présence des complexités telles que l'effet Faraday [57][86] ou la phase nématique [87]. Nous allons nous intéresser ici aux conséquences de l'introduction de deux nouveaux types de complexité dans la théorie microscopique de la diffusion multiple de la lumière : l'activité optique naturelle et l'effet magnéto-chiral.

Dans un premier temps, nous considérerons le transport de la lumière dans un milieu où la symétrie miroir est absente, ce que nous permet de traiter l'activité optique naturelle en diffusion multiple. Il est important de remarquer que ce travail constitue, à notre connaissance, à la première étude microscopique de la diffusion multiple de la lumière dans les milieux chiraux. En utilisant cette approche microscopique, nous allons introduire le libre parcours moyen de transport chiral ℓ_C^* , que nous allons calculer pour quelques systèmes chiraux simples. En outre, nous montrerons que ℓ_C^* est associé à la polarisation résiduelle de la lumière diffuse dans les milieux chiraux, qui a été observée dans les simulations numériques de Ablitt *et al.* [88].

Dans un deuxième temps, nous allons ajouter un nouvel ingrédient à la théorie : le champ magnétique externe $\mathbf{B}$, responsable pour la brisure de la symétrie du renversement temporel. La présence de $\mathbf{B}$ nous permet de développer une théorie microscopique pour l'effet magnéto-chiral en diffusion multiple. Ce genre de milieu possède des propriétés de symétrie très particulières (à savoir, la brisure simultanée des symétries miroir et du renversement temporel) qui permettent *a priori* l'existence d'un « super » courant photonique, c'est-à-dire un courant qui persiste en l'absence d'un gradient spatial de la densité photonique. Nous allons donc spéculer sur l'existence de ce courant. Enfin, nous considérerons les propriétés de la lumière transmise à travers d'une tranche magnéto-chirale et nous démontrerons l'existence d'une magnéto-transmission *linéaire*.

Transport de la lumière dans les milieux chiraux

Libre parcours moyen de transport chiral

Afin de traiter la chiralité en diffusion multiple, nous allons employer un modèle similaire à celui présenté au chapitre 4 pour décrire les milieux magnéto-chiraux inhomogènes : diffuseurs ponctuels placés dans des configurations géométriques chirales.

Comme nous en avons discuté au chapitre 3, une des quantités fondamentales dans la théorie microscopique de transport est le tenseur d'intensité spécifique, dont les composants I_{ij} décrivent l'état de polarisation de la radiation diffusée dans la direction $\mathbf{p}$. L'intensité spécifique peut être écrite ainsi [60] :

$$\mathbf{I}_{\mathbf{p}}(\mathbf{q}, \chi) = i [\mathbf{G}_{\mathbf{p}} - \mathbf{G}_{\mathbf{p}}^*] - i \mathbf{G}_{\mathbf{p}} \cdot \Gamma_{\mathbf{p}}(\mathbf{q}, \chi) \cdot \mathbf{G}_{\mathbf{p}}^*, \quad (5.1)$$

où $\mathbf{q}$ est la transformé de Fourier du vecteur position et $\mathbf{G}_{\mathbf{p}}$ le tenseur de Dyson-Green introduit au chapitre 3. Nous avons défini un pseudo-scalaire χ qui est associé à la géométrie chirale des diffuseurs. Le tenseur $\Gamma_{\mathbf{p}}(\mathbf{q}, \chi)$ décrit la faible dépendance angulaire de la radiation diffuse et qui obéit à l'équation intégrale :

$$\Gamma_{\mathbf{p}}(\mathbf{q}, \chi) = \mathbf{L}_{\mathbf{p}}(\mathbf{q}) + \sum_{\mathbf{p}'} \Gamma_{\mathbf{p}'}(\mathbf{q}, \chi) \cdot \mathbf{G}_{\mathbf{p}} \otimes \mathbf{G}_{\mathbf{p}'}^* \cdot \mathbf{U}_{\mathbf{p}\mathbf{p}'} \quad (5.2)$$

où $\mathbf{U}_{\mathbf{p}\mathbf{p}'}(\mathbf{q}, \chi)$ est le vertex irréductible défini par (3.27). Le tenseur courant est défini comme $\mathbf{L}_p(\mathbf{q}) \equiv 2(\mathbf{p} \cdot \mathbf{q})\mathbf{U} - \mathbf{p}\mathbf{q} - \mathbf{q}\mathbf{p}$ (où $\mathbf{U}$ est le tenseur d'unité). Dans un milieu isotrope et achiral, le résultat classique de Boltzmann s'applique : $\Gamma_{ij\mathbf{p}}(\mathbf{q}) = (\mathbf{p} \cdot \mathbf{q})\gamma_0\delta_{ij}$ où $\gamma_0 = 2/(1 - \langle \cos \theta \rangle)$.

Notre but sera de résoudre explicitement l'Éq. (5.2) pour trouver une expression pour le libre parcours moyen de transport chiral ℓ_C^* . Pour cela, nous allons faire une seule approximation : l'approximation de Boltzmann (voir Eq. (3.16)), ce qui signifie que nous ne considérons que les faibles densités des diffuseurs. En utilisant cette approximation, il est possible montrer que l'intensité spécifique (5.1) dans des milieux chiraux s'écrit ainsi :

$$\mathbf{I}_{\mathbf{p}} \sim \Delta_{\mathbf{p}} + \frac{i}{2}\ell_s(\hat{\mathbf{p}} \cdot \mathbf{q}) \left[\left(\frac{\gamma_0 + \xi\gamma_C}{1 + \xi^2} \right) \Delta_{\mathbf{p}} + \left(\frac{\gamma_C - \xi\gamma_0}{1 + \xi^2} \right) \Phi_{\hat{\mathbf{p}}} \right], \quad (5.3)$$

où $(\Delta_{\mathbf{p}})_{ij} \equiv \delta_{ij} - p_i p_j / p^2$ et $\Phi_{ij} = i\epsilon_{ijk}\hat{p}_k$ sont deux tenseurs transverses, et ℓ_s est le libre parcours moyen conventionnel. Les pseudo-scalaires ξ et γ_C sont associés, respectivement, au pouvoir rotatoire et à la géométrie chirale du milieu. Notons que l'équation (5.3) suggère la définition d'un libre parcours moyen de transport ℓ^* :

$$\ell^* \equiv \frac{\ell_s}{2} \left(\frac{\gamma_0 + \xi\gamma_C}{1 + \xi^2} \right), \quad (5.4)$$

et, de la même façon, d'un libre parcours moyen de transport chiral ℓ_C^* :

$$\ell_C^* \equiv \frac{\ell_s}{2} \left(\frac{\gamma_C - \xi\gamma_0}{1 + \xi^2} \right). \quad (5.5)$$

En l'absence de chiralité (i.e., pour $\xi = \gamma_C = 0$), ℓ^* se réduit au libre parcours moyen de transport ordinaire $\ell_s/(1 - \langle \cos \theta \rangle)$ et $\ell_C^* = 0$. A notre connaissance, le libre parcours moyen de transport chiral ℓ_C^* n'a jamais été introduit auparavant.

Nous avons calculé numériquement ℓ_C^* pour des différentes configurations de diffuseurs ponctuels. Nous avons vérifié que ℓ_C^* s'annule toujours pour les géométries achirales, e.g., pour des systèmes composés d'un, deux ou trois diffuseurs. Nous avons également évalué ℓ_C^* pour des systèmes chiraux simples, comme par exemple le « twisted H » et la hélice. Pour le « twisted H » ℓ_C^* s'annule quand l'angle α entre les bras du « twisted H » vaut $\alpha = n\pi/2$ (où n est un entier). En outre, nous avons montré que ℓ_C^* suit la même dépendance synodale

$\psi \sim \sin(2\alpha)$ du « paramètre d'ordre chiral » ψ proposé par Harris *et al.* [75][76] [77], ainsi que les propriétés de diffusion des milieux magnéto-chiraux (voir chapitre 4). Ces faits démontrent que les propriétés du transport de la lumière dans les milieux inhomogènes sont aussi sensibles au degré de la chiralité ψ du milieu.

Polarisation circulaire de la lumière diffuse

Dans ce paragraphe nous allons utiliser les résultats de la section précédente pour analyser le rôle physique de ℓ_C^* dans le processus de diffusion multiple de la lumière par une tranche de longueur L . En utilisant l'Éq. (5.3) à la tranche et en appliquant les conditions radiatives aux bords [68], nous obtenons :

$$I_\sigma(\theta, L) = \frac{4}{3} \frac{\ell^*}{L + \frac{4}{3}\ell^*} \left[1 + \frac{3}{2} \cos \theta \left(1 + \frac{\ell_C^*}{\ell^*} \sigma \right) \right], \quad (5.6)$$

où $\sigma = \pm 1$ représente l'état de polarisation circulaire de la lumière et θ dénote l'angle entre le vecteur d'onde émergent et la normale de la tranche. L'équation (5.6) implique que la radiation diffuse possède *un degré résiduel de polarisation circulaire*. La polarisation circulaire est quantifiée par le paramètre de Stokes $V(\theta) = I_+(\theta) - I_-(\theta)$:

$$\frac{V(\theta)}{I(\theta)} = \frac{\frac{3}{2} \cos \theta}{1 + \frac{3}{2} \cos \theta} \left(\frac{\ell_C^*}{\ell^*} \right) \quad (5.7)$$

où $I(\theta) = I_+(\theta) + I_-(\theta)$. Notre résultat microscopique (5.7) est en accord avec des simulations de Monte Carlo [88], qui ont montré que la lumière diffuse par un milieu chiral inhomogène acquiert un degré de polarisation circulaire, un fait qui peut avoir des applications potentielles en imagerie [88]. Ce résultat est impossible dans un milieu achiral où la lumière émerge complètement dépolarisée suite aux multiples diffusions.

Transport de la lumière dans les milieux magnéto-chiraux

Transmission magnéto-chirale

Motivés par la récente observation d'une *magnéto-transmission quadratique* dans les milieux Faraday actifs et achiraux [89], nous allons étudier la dépendance en champ magnétique $\mathbf{B}$ du

coefficient de transmission $T(\theta, B)$ d'un milieu chiral. Pour cela, nous emploierons la théorie vectorielle de transport de la lumière, en prenant en compte la dépendance en $\mathbf{B}$ des différentes quantités de transport. Le coefficient de transmission pour une tranche de longueur L est obtenue en évaluant l'intensité spécifique (5.3) à la sortie $z = L$ du faisceau lumineux. Pour $\mathbf{B}$ appliqué au long de la normale de la tranche, nous prédisons la présence d'une *magnéto-transmission linéaire* (normalisée) qui possède la forme suivante :

$$\frac{\delta T(\theta, B)}{T(\theta)} = \frac{3}{4} \frac{\ell_{MC}^*}{\ell^*} \frac{\cos^2 \theta - (1/2)}{1 + (3/2) \cos \theta}. \quad (5.8)$$

Nous avons introduit le libre parcours moyen de transport « magnéto-chiral » $\ell_{MC}^* \equiv \gamma_{MC} B \ell_s$. Typiquement, la magnéto-transmission linéaire est de l'ordre de $\gamma_{MC} B \sim (VB/\Gamma k^2) \ell_C^*/\ell^*$ et nous pouvons envisager de la mesurer dans les solutions des molécules Faraday actives dispersées.

Existe-t-il un « super » courant photonique dans les milieux magnéto-chiraux inhomogènes ?

Dans les milieux magnéto-chiraux, où les symétries miroir et du renversement du temps sont simultanément brisées, un courant du type $\mathbf{J}_C(\mathbf{r}, t) = D_C \mathbf{B} \nu(\mathbf{r}, t)$ est permis par symétrie (où $\nu(\mathbf{r}, t)$ est la densité d'énergie électromagnétique). Soulignons qu'il s'agit bien d'un courant à l'absence d'un gradient spatial d'énergie. Par conséquent, il faut que la quantité D_C soit un *pseudo-scalaire* pour qu'elle puisse remplacer le rôle du gradient spatial dans la loi de Fick, $\mathbf{J}(\mathbf{r}, t) = -\mathbf{D}(\mathbf{B}) \cdot \nabla \nu(\mathbf{r}, t)$. Ce « super » courant $\mathbf{J}_C$ serait alors une autre manifestation macroscopique de la symétrie de parité microscopiquement brisée, ainsi que des propriétés de transport dans les milieux chiraux et la diffusion magnéto-chirale, comme nous l'avons vu dans la discussion précédente. Il est intéressant de remarquer que le « super » courant a formellement la même structure que le courant de London dans un supraconducteur, $\mathbf{J}_L = e \mathbf{A} \nu$, où $\mathbf{J}_L$ et $\mathbf{A}$ sont des vecteurs impairs sous parité. $\mathbf{J}_L$ s'impose par invariance de jauge, contrairement au courant électronique ordinaire (Ohmique) qui est décrit par la loi de Fick et qui obéit à l'invariance de jauge. Pour le courant magnéto-chiral $\mathbf{J}_C$ cette contrainte de jauge n'existe pas car elle implique l'observable $\mathbf{B}$. Nous avons dérivé une expression microscopique pour $\mathbf{J}_C$ et

nous l'avons calculée en utilisant notre modèle pour l'effet magnéto-chiral, présenté au chapitre 4. Malheureusement, nous sommes arrivés à la conclusion que, au moins pour ce modèle, le « super » courant s'annule. Nous suspectons qu'il doit exister un fort argument de symétrie ou quelque argument basé sur la théorie des groupes capable d'expliquer ce résultat. Cependant, il nous échappe pour l'instant.

Part II

Statistics of resonance widths in disordered optical media

Plan of part II

This part of the thesis is devoted to the study of the statistical properties of resonance widths in *open multiple scattering systems* composed of point-dipoles. We will focus on two different aspects of the distribution of resonance widths $P(\Gamma)$. First, we will analyze how the phenomenon of Anderson localization manifests itself in $P(\Gamma)$ for one-, two- and three-dimensional disordered systems using a scalar treatment for wave propagation and scattering. By means of this analysis we conclude that $P(\Gamma)$ exhibits an universal power law decay $P(\Gamma) \sim 1/\Gamma$ in the localized regime, which we suggest to be an unambiguous manifestation of Anderson localization. Second, we will study the impact of the presence of an external magnetic field in $P(\Gamma)$ for magneto-optical random media in the diffusive regime. A possible application to random lasers will be discussed. This second part is organized as follows. In Chapter 6, the general concepts and properties of $P(\Gamma)$ in open mesoscopic systems are discussed. Chapter 7 is dedicated to the phenomenon of Anderson localization, where the main localization criteria will be presented. In Chapter 8, recent results concerning $P(\Gamma)$ in situations related to the Anderson transition will be considered, for the localized regime, for the extended regime and around the critical point. In Chapter 9 the results concerning $P(\Gamma)$ in one-, two- and three-dimensions in the localized regime will be presented. Finally, Chapter 10 is devoted to the analysis of $P(\Gamma)$ in a magnetic field and also contains an outline for future work.

Plan de la partie II

La deuxième partie de la thèse est consacrée à l'étude des propriétés statistiques des largeurs de résonance dans les *milieux diffuseurs désordonnés ouverts*. Nous nous intéresserons à deux aspects différents de la distribution des largeurs de résonance $P(\Gamma)$. Dans un premier temps, nous allons analyser comment le phénomène de la localisation d'Anderson se manifeste en $P(\Gamma)$ pour les systèmes désordonnés uni-, bi- et tridimensionnels en utilisant une approche scalaire pour la propagation et la diffusion ondulatoire. Cette analyse nous permet de conclure que $P(\Gamma)$ suit une loi de puissance universelle $P(\Gamma) \sim 1/\Gamma$, un comportement que nous interprétons comme une manifestation non équivoque de la localisation d'Anderson. Dans un deuxième temps, nous allons étudier l'impact de la présence d'un champ magnétique externe en $P(\Gamma)$ pour systèmes magnéto-optiques aléatoires en régime diffus. Une application possible aux lasers aléatoires sera également discutée. La deuxième partie de la thèse est organisée de la manière suivante. Au Chapitre 6 les propriétés et concepts généraux de $P(\Gamma)$ dans les systèmes mésoscopiques ouverts seront présentés. Le Chapitre 7 est dédié au phénomène de la localisation d'Anderson, où nous discuterons les principaux critères pour la localisation. Au Chapitre 8 nous analyserons les résultats récents concernant la distribution $P(\Gamma)$ pour les situations liées à la transition d'Anderson, le régime localisé, le régime diffus et autour du point critique. Au Chapitre 9 nous présenterons les résultats concernant la distribution $P(\Gamma)$ à une, deux et trois dimensions dans le régime localisé pour les milieux diffuseurs désordonnés ouverts composés de dipôles ponctuels. Finalement, le Chapitre 10 est consacré à l'analyse de $P(\Gamma)$ sous l'influence du champ magnétique. Il contient également des perspectives et des propositions pour un travail futur.

Chapter 6

Decay rate statistics in open mesoscopic systems

6.1 Introduction

6.1.1 Mesoscopic wave transport

Wave transport in a disordered system is generally said to be in the *mesoscopic* regime if the system size L obeys the following inequalities:

$$\ell < L < L_\phi. \tag{6.1}$$

The first inequality compares L to the *mean free path* ℓ , the typical distance between two scattering events (e.g. by impurities or defects inside the sample), ensuring that disorder is sufficiently strong to generate multiple scattering. If $L \ll \ell$ the disorder is weak, the wave is hardly scattered and wave propagation is ballistic. The second inequality compares L to the so-called *coherence length* L_ϕ , a distance beyond which the phase coherence in wave propagation is lost. It should be emphasized that disorder, associated with the first inequality, does *not* destroy the phase, but just scrambles it. For quantum mechanical waves (e.g. electrons), the phase of the wavefunction is completely destroyed by inelastic processes, among which the electron-phonon scattering and the electron-electron interaction. In order to observe manifestations of phase-coherence in electronic transport, such as quantum interferences, one has to cool

down the system: L_ϕ is typically of the order of microns at very low temperatures. For classical waves (electromagnetic, elastic or acoustic waves), propagation is generally phase-coherent though limited by absorption. An important exception of phase-coherent wave transport is light propagation in disordered atomic media, where the Doppler effect induces dephasing (inhomogeneous line broadening). Phase-coherence constitutes the main characteristic of mesoscopic wave transport and is at the origin of several fascinating physical phenomena, such as universal conductance fluctuations [90].

6.1.2 Wave transport in open mesoscopic systems: the concept of resonances

It is important for many experimental situations in mesoscopic wave transport to consider *open* media, i.e., media where waves can “leak” through the sample boundaries. In an open system, the eigenstates are not “bound states” like in an (ideally) closed system but have become *resonances* with a finite energy width Γ or, equivalently, with a finite lifetime $t \sim 1/\Gamma$.

The properties of resonances are not only fundamental for wave dynamics in open mesoscopic systems, but also they are directly related to quantities measurable in experiments. In electronic systems, in particular, it is known that the conductance can be expressed in terms of the resonances of a quantum dot in the regime of Coulomb blockade [91]. In optical systems, the most important application is associated with the investigation of random lasers [92][93]. In a random laser, the feedback necessary for lasing is not provided by mirrors, as in ordinary lasers, but by long light paths in a disordered medium, along which amplification occurs. The condition for lasing is achieved when the gain due to amplification becomes larger than the decay caused by leakage through the system boundaries. As a result, the laser threshold is in principle determined by the smallest decay rate among the “eigenmodes” or resonances [94][95]. This intuitive conclusion is confirmed by a complete quantum mechanical treatment [96].

The treatment of wave transport in mesoscopic systems generally tries to avoid microscopic details of the system (e.g. the configuration of impurities in a semiconductor, scatterers in an optical sample or the precise shape of the system boundaries), which usually can not be taken into account exactly. A statistical approach is proposed, in which one considers an ensemble of different microscopic configurations with the same macroscopic parameters. Mesoscopic wave transport often deals with the statistical analysis of leakage-related quantities, such as

transmission coefficients, resonance widths and time-delays [90][97].

6.2 Statistical description of open mesoscopic systems

Resonances in open media show up as poles of the so-called scattering (energy-dependent) S -matrix, which relates the outgoing scattered complex wave amplitudes to the incoming wave amplitudes. The S -matrix can be expressed in terms of the total Hamiltonian of the open system $\mathcal{H}_{tot}$ as [97]:

$$S = I - 2\pi i W^\dagger \frac{1}{\mathcal{E} - \mathcal{H}_{tot}} W, \quad (6.2)$$

where I is the unit matrix and W is an operator that describes the coupling between the system and the environment, generally modelled as attached leads to its ends. Flux conservation in wave transport imposes the unitarity of the S -matrix, i.e., $SS^\dagger = 1$ [50]. The total Hamiltonian of the open system $\mathcal{H}_{tot}$ relates to the one of its closed equivalent $\mathcal{H}_{clo}$ according to:

$$\mathcal{H}_{tot} = \mathcal{H}_{clo} - i\pi W^\dagger W. \quad (6.3)$$

From Eq. (6.2) one sees that the eigenvalues of $\mathcal{H}_{tot}$ are the poles of the S -matrix. Notice that, due to the opening of the system, $\mathcal{H}_{tot}$ in (6.3) is non-Hermitian. As a result, its eigenvalues are complex-valued: $\mathcal{E}_n = E_n - \frac{i}{2}\Gamma_n$, where E_n and Γ_n are called position and width of the n -nth resonance, respectively.

The knowledge of the S matrix, including its energy dependence, facilitates in principle the complete description of the scattering problem. Another important quantity to describe open mesoscopic systems is the delay time $\tau(\mathcal{E})$. The delay time $\tau(\mathcal{E})$, studied originally by Eisenbud and Wigner [98], physically represents the typical time the wave remains in the scattering region. As well as resonance widths, it can be expressed in terms of the S -matrix [97]:

$$\tau(\mathcal{E}) = i\text{Tr} \frac{\partial S^\dagger}{\partial \mathcal{E}} S, \quad (6.4)$$

which is real-valued since S is a unitary operator.

For a detailed and pedagogical discussion about delay time, not only for mesoscopic wave

transport but also for several other physical applications, to reader is referred to the recent review paper by de Carvalho and Nussenzveig [99].

There are essentially three different theoretical tools to investigate the statistical properties of the S matrix, allowing the calculation of resonance widths, phase shifts and time delays: the semiclassical approach, the stochastic approach, and “alternative methods”, which will be discussed in the following.

6.2.1 Stochastic approach: Random matrix theory

As was anticipated in the introduction, the fact that mesoscopic systems with the same macroscopic parameters can differ in several microscopic details suggests the employment of a statistical description of the problem of wave transport and scattering. This is the very basis underlying *Random Matrix Theory* (RMT) [100]. In RMT, one completely neglects many microscopic details of the system, taking into account solely its global properties and symmetries, which manifest themselves in the system Hamiltonian. In RMT, one considers an ensemble of random Hamiltonians determined only by the global symmetries of the system. For example, if the system does not exhibit any particular symmetry, one considers the set of all possible Hermitian matrices, called the Gaussian Unitary Ensemble (GUE). If the system is time-reversal invariant, the matrices must be real and symmetric, a set referred to as Gaussian Orthogonal Ensemble (GOE). The main advantage of RMT can be attributed to its independency on the microscopic details of the system. In this sense, RMT can be regarded as universal and it turns out to be an efficient mathematical tool for the study of disordered mesoscopic systems¹. The original RMT only applies to closed systems, i.e., it is valid solely for $\mathcal{H}_{clo}$ only. The actual open systems are often treated *a posteriori*, in the same spirit of (6.3). For details on the methods developed to adjust RMT to open systems, we refer to Ref. [97].

RMT, as originally conceived in the 1960’s by Wigner, Dyson and Metha in the context of nuclear physics [102][100], does not only apply to mesoscopic systems, but also to several other domains of physics. In the 1980’s, Bohigas [103] surmised that RMT could be used as tool to describe the statistical spectra of quantum chaotic systems, that is, quantum systems

¹To be precise, RMT predictions are actually universal provided the number of propagating channels tends towards infinity (see Ref. [101]).

whose classical analogs exhibit chaos [104]. Numerous subsequent studies have confirmed the utility of RMT for quantum chaotic systems, but an analytical proof to justify its applicability in this domain remains unknown, despite some recent efforts in this sense [105]. For disordered systems, the application of RMT was first conjectured by Gor’kov and Eliashberg [106] and was then proven by Efetov using the so-called non linear σ -model [107]. For a very complete and detailed discussion of RMT in mesoscopic systems, including several applications, the reader is referred to the review paper by Beenakker [101].

As already mentioned, the remarkable success of RMT in several domains is mainly due to its universal character. At the same time, this universality implies that RMT neglects many specific details of the system under study. Consequently, RMT is expected to give correct predictions only in the limiting case where all physical scales can be regarded as irrelevant. Probably the most important physical situation where this is certainly not true, and hence where standard RMT is expected to fail, is the regime of Anderson localization. Anderson localization, which will be discussed in more detail in Chapter 7, refers to an inhibition of wave diffusion due to destructive interference of multiple scattered waves. The onset of Anderson localization is associated with a typical length scale - the localization length, which measures the spatial extent of localization - making the use of the standard “Wigner-Dyson” RMT inviable [101]. Further developments in RMT, where the distribution of scattering matrices is obtained from the Dorokhov-Mello-Pereyra-Kumar (DMPK) equation (often called DMPK approach to RMT), have extended its validity for wave transport in the localized regime [101]. However, both the “Wigner-Dyson” and the DMPK approaches to RMT are restricted to one-dimensional and quasi-one-dimensional systems, which still constitutes the main limitation of RMT.

6.2.2 Semiclassical approach

The semiclassical approach to analyze the statistical properties of the S -matrix applies to quantum systems which are chaotic in the classical limit. This is an important conclusion in quantum chaos (for a review, see Ref. [108]). The starting point of the semiclassical approach is to represent the quantum S -matrix elements in terms of a sum over the orbits of the corresponding classical system [109]. This can be accomplished by considering the so-called quantum graphs to model quantum scattering systems. By attaching infinite leads at the nodes of these graphs,

it has been demonstrated that they can model scattering systems exhibiting chaotic scattering. This mathematical tool allows to explicitly write the quantum S -matrix, and to efficiently calculate the resonances, as well as the time delays [110]. The results obtained for the statistical properties of the S -matrix agree with the predictions of RMT and serve to provide a deeper understanding of the connection between RMT for quantum systems and their underlying classical chaotic dynamics. For a detailed reference about quantum graphs, including the comparisons with RMT predictions, see Ref. [111] .

6.2.3 Alternative approaches

The statistical study of wave propagation in disordered open media can be carried out by diagonalizing large complex matrices corresponding to the total Hamiltonian of the system within a given physical model, such as the Anderson model or the kicked rotor model. As in the case of RMT, the opening of the system for such models is treated *a posteriori*, and is generally considered as a perturbation of the corresponding closed system. Another important alternative approach is the recently implemented algorithm capable to extract resonance positions and widths with high precision, using the spectral analysis of time correlation for dissipative dynamical systems. This method has been applied to compute unimolecular decay rates [112].

The main disadvantage of these “alternative” methods is the fact that they are usually very time-consuming at a computational level, since they involve the diagonalization of large complex matrices. However, they constitute the only alternative to situations where RMT does not apply, such as systems with high dimensionality (two- and three- dimensions). In Chapter 9 the distribution of resonance widths in the localized regime will be investigated in one-, two- and three- dimensional systems using a new “alternative” approach, namely the point-dipole model to describe light propagation in disordered media, an *ab initio* approach introduced by Rusek and Orłowski [81][82].

Résumé du chapitre 6 : Statistique des largeurs de résonance dans les milieux mésoscopiques ouverts

Le transport ondulatoire dans les milieux désordonnés est dit dans le régime *mésoscopique* quand la taille L du système satisfait les inégalités suivantes:

$$\ell < L < L_\phi, \tag{6.5}$$

où ℓ est le libre parcours moyen, la distance typique entre deux diffuseurs, et L_ϕ est la longueur de cohérence du système, la distance au delà de laquelle la cohérence de phase de l'onde est détruite. La cohérence de phase est la caractéristique principale du transport mésoscopique.

Dans un grand nombre des situations expérimentales de transport mésoscopique, il est important de considérer les milieux ouverts, i.e. les systèmes où les ondes peuvent «fuir» à travers de ses bordes. Dans les milieux ouverts, les « états propres » du système ne sont pas états liés comme dans les milieux (idéalement) fermés, mais ils deviennent *résonances* avec une largeur finie en énergie Γ , c'est à dire un temps de vie fini $t \sim 1/\Gamma$. Dans la deuxième partie de la thèse, nous nous intéressons au transport mésoscopique des ondes dans les milieux ouverts en utilisant une *approche statistique*.

Description statistique du transport ondulatoire dans les systèmes mésoscopiques ouverts

Les résonances en milieux ouverts se manifestent comme les pôles de la matrice S de diffusion, qui relie les amplitudes des ondes incidente et diffusée. La matrice S peut être exprimée en fonction de l'Hamiltonien total $\mathcal{H}_{tot}$ du système ouvert [97] :

$$S = I - 2\pi i W^\dagger \frac{1}{\mathcal{E} - \mathcal{H}_{tot}} W, \quad (6.6)$$

où I est la matrice identité et W est l'opérateur qui décrit le couplage entre le système et ses environnements. L'Hamiltonien total du système $\mathcal{H}_{tot}$ s'exprime en fonction de celui de son équivalent fermé comme

$$\mathcal{H}_{tot} = \mathcal{H}_{clo} - i\pi W^\dagger W. \quad (6.7)$$

En raison de l'ouverture du système, $\mathcal{H}_{tot}$ n'est pas hermitique. Par conséquent, ses valeurs propres sont complexes : $\mathcal{E}_n = E_n - \frac{i}{2}\Gamma_n$, où E_n et Γ_n sont la position et la largeur de la n -ième résonance, respectivement.

Il y a généralement trois manières d'étudier les propriétés statistiques de la matrice S , nous permettant de calculer les largeurs de résonance : l'approche stochastique, l'approche semiclassique et les « méthodes alternatives ».

L'approche stochastique repose sur la Théorie de Matrices Aléatoires (TMA) [101]. L'idée centrale de la TMA est de négliger un grand nombre de détails microscopiques du système, en ne considérant que ses propriétés globales et ses symétries. Ainsi, la TMA considère un ensemble d'Hamiltoniens aléatoires déterminés seulement par les symétries globales du système (e.g. symétrie de renversement du temps). Le principal avantage de la TMA est justement sa généralité, qui vient de l'indépendance des détails microscopiques du système. D'une autre côté, la TMA ne s'applique qu'aux systèmes de dimensionnalité réduite (1D et quasi 1D), ceci constitue sa principale faiblesse.

L'approche semiclassique s'applique aux systèmes quantiques qui sont chaotiques dans la limite classique, les systèmes classiquement chaotiques [108]. L'idée de l'approche semiclassique

est de représenter les éléments de la matrice S quantique en fonction des orbites classiques correspondantes [109]. Cette idée peut être réalisée en considérant un outil mathématique - les graphes quantiques [111] - qui nous permet d'écrire explicitement la matrice S quantique et de calculer les résonances [110].

Finalement, les « approches alternatives » sont basées sur la diagonalisation de large matrices complexes qui correspondent à l'Hamiltonien total du système en utilisant un modèle physique donnée (comme le modèle d'Anderson ou le modèle kicked rotor). Le désavantage majeur de ce genre d'approches est qu'en général il requiert beaucoup de temps de calcul numérique. Cependant, il est fréquemment la seule alternative pour calculer la matrice S et les résonances dans les cas où la TMA ne s'applique pas, comme les systèmes de haute dimensionnalité (2D et 3D). Au chapitre 9 nous étudierons la distribution de largeurs de résonance au régime localisé en utilisant une nouvelle « approche alternative » : le modèle des diffuseurs ponctuels pour décrire la propagation des ondes en milieux désordonnés, introduit par Rusek et Orłowski [81][82].

Chapter 7

Localization of waves

After the 1958 pioneer paper by Anderson [10], the subject of wave localization and metal-insulator transitions became an extremely active topic of research. The aim of the present chapter is to briefly overview the phenomenon of Anderson (or strong) localization of waves in disordered media, focusing mainly in the fundamental concepts and definitions. For a more detailed and complete description, the reader is referred to review papers [113][114][115][116][117]. The optical counterpart of strong electronic localization will also be discussed.

7.1 The concept of wave localization: Anderson model

Localization, as proposed by Anderson, is defined as an inhibition of wave diffusion in infinite disordered media. In his original paper, Anderson aimed to understand the electronic transport properties in doped semiconductors, and in particular the interplay between disorder and electronic diffusion [10]. He formulated a model to describe the hopping of tightly bound (but still mobile) electrons from different impurity sites, for which the so-called Anderson Hamiltonian with diagonal disorder has the form

$$\mathcal{H} = \sum_n \epsilon_n |n\rangle \langle n| + \sum_{n,m} V |n\rangle \langle m|. \quad (7.1)$$

In Eq. (7.1), which probably represents the simplest model capable to describe localization effects, the disorder is taken into account by assuming that the energy ϵ_n of the n -th impurity

site is a random variable. The summation in the second term is usually performed over the nearest neighbors only and V is a deterministic hopping matrix element. Using this model, Anderson has demonstrated that, given an electron initially at a given site n_0 , the population of the other sites rapidly decays with the distance to the site n_0 after an infinite time, provided the disorder (i.e., the concentration of impurities) is sufficiently strong.

The physical origin of this “absence of diffusion” is due to interference effects between multiple scattered waves in the random medium. On a short length scale, interference effects cause fluctuations in the amplitude of the eigenfunctions $\psi(\mathbf{r})$, reducing the diffusion constant, but at very large distances they can eventually induce eigenfunctions to decay to zero. This decay is exponential

$$|\psi(\mathbf{r})| \sim \exp(-|\mathbf{r} - \mathbf{r}_0|/\xi), \quad (7.2)$$

and is characterized by the localization length ξ , which measures the spatial extent of localization. Such a decay is believed to lead to a vanishing diffusion constant in *infinite* systems, although some pathological situations do exist to suggest that this is not always true [118].

Although originally formulated in the realm of electronic transport, the phenomenon of Anderson localization, being essentially an ondulatory phenomenon, is expected to occur for any kind of wave. For light¹, in particular, the observation of Anderson localization [119] has been subject to an intense controversy and is still lively debated [120][121].

It should be emphasized that these previous definitions of localization are only valid for *infinite* random media. It turns out that they are of limited utility in practice, since actual experiments are, at least in three dimensions, generally performed in *finite* and *open* media. Therefore, the observation of Anderson localization and, consequently, the formulation of a criterion to determine its onset, should be related to manifestations of leakage in observable quantities. This notion constitutes the underlying basis of the Thouless criterion for localization, that will be discussed in the following section.

¹See section 7.3.

7.2 Main localization criteria

7.2.1 Thouless criterion

In an open medium, the “eigenfunctions” are resonances with a finite energy width Γ (or, equivalently, a finite lifetime $t \sim 1/\Gamma$)² due to leakage. The Thouless criterion [113] is based upon the comparison of two fundamental time scales: the Thouless time, t_T , and the Heisenberg time, t_H . The Thouless time corresponds to the typical time an excitation requires to traverse the whole system (size L), $t_T \sim L^2/D$, with the D diffusion constant. The Heisenberg time characterizes the maximal time scale in the system, $t_H \sim 1/\Delta E$, with ΔE the mean level spacing. The Thouless criterion asserts that localization can be said to occur when $t_T > t_H$, that is incompatible with the diffusion process for which t_T must be smaller than t_H .

The Thouless criterion can also be rephrased in terms of energy scales, if one compares the typical energy width $\Gamma \sim D/L^2$ to the mean level spacing $\Delta E \sim 1/t_H$. Recalling that $\Delta E \approx 1/\rho(E)L^d$, with $\rho(E)$ the density of states per unit of volume and d the dimensionality of the system, and introducing the “dimensionless conductance” $g \equiv \Gamma/\Delta E$, one concludes that the Thouless criterion is equivalent to

$$g \sim \rho(E)D(E)L^{d-2} < 1. \quad (7.3)$$

Physically, Eq. (7.3) means that the energy width corresponding to the leaky wavepacket associated with the diffusion process contains only one “eigenstate”. As a result, diffusion cannot occur for $g < 1$ and the wave must be localized. On the other hand, $g > 1$ corresponds to the extended regime. The critical situation is expected to occur for $g \approx 1$.

It is instructive to view the Thouless criterion in a more mesoscopic context. Mesoscopic wave transport deals essentially with transmission-related quantities. For a sample of length L and N transverse modes, important observational parameters are the transmission coefficient for an incoming mode a into an outgoing mode b ³, T_{ab} , the total transmission for incoming mode a , $T_a = \sum_b T_{ab} \sim \ell^*/L$, and the total transmittance, $T = \sum_{ab} T_{ab} \sim N\ell^*/L$, with ℓ^* the transport mean free path. The total transmittance T is, by the Landauer formula [90],

²For quantum waves, we assume that $\hbar = 1$. For classical waves, Γ should be regarded as a modal frequency.

³The modes or “channels” can be, for instance, the incoming and outgoing directions.

equivalent to the conductance G in electronic systems

$$T = \frac{h}{e^2} G. \quad (7.4)$$

The ensemble-averaged dimensionless conductance is thus

$$g = \langle T \rangle \sim N\ell^*/L \quad (7.5)$$

Using Eq. (7.5) and the fact that the localization length ξ in a mesoscopic sample is defined by $\xi \approx N\ell^*$, one deduces that the Thouless criterion [Eq. (7.3)] is equivalent to

$$L > \xi, \quad (7.6)$$

showing that the size of the system should be larger than the localization length in the localized regime.

7.2.2 Ioffe-Regel criterion

Ioffe and Regel formulated a qualitative criterion for the localization transition in disordered media based on the comparison between the wavelength λ and the mean free path ℓ . The Ioffe-Regel criterion states that, in three dimensions, localization sets in when λ is of the order of ℓ [114]:

$$k\ell < \text{constant} \approx 1, \quad (7.7)$$

where $k = 2\pi/\lambda$ is the wavenumber *inside* the medium. It is important to stress that the mean free path featuring in (7.7) is the extinction mean free path, defined as the coherence length of the incident beam⁴ and *not* the transport mean free path ℓ^* .

The constant in (7.7) is model-dependent. For the three dimensional Anderson model, an analytical calculation based in the so-called potential-well analogy gives $k\ell \lesssim 0.84$ as the threshold for the onset of the localized regime [122]. The potential-well analogy is based on the

⁴See Eq. (3.8).

fact that the localization problem (or, more precisely, the self-consistent approximation to it) can be mapped onto the one of a particle being scattered or trapped by a local potential well (see, e.g., Ref. [123]).

7.2.3 Scaling approach for Anderson localization

Another important achievement in localization theory is the scaling approach, developed by Abrahams *et al.* [124]. The main objective of the scaling theory of localization is understanding the behavior of the conductance g as a function of the system size L . When the disorder is weak but strong enough to generate multiple scattering (i.e., for $L \gg \ell$), the wavefunctions are extended and the conductance $g(L)$ is given by the Ohm law:

$$g(L) = \sigma L^{d-2}, \quad (7.8)$$

where Eq. (7.3) and the Einstein relation $\sigma \sim \rho D$ were used (with σ the conductivity). On the other hand, if the disorder is sufficiently strong to exponentially localize the wavefunctions, $g(L)$ is given by

$$g(L) = \exp(-L/\xi), \quad (7.9)$$

with ξ the localization length and $L > \xi$.

Knowing the behavior of the conductance $g(L)$ in the diffusive regime - Eq. (7.8) - and in the localized regime - Eq. (7.9) - Abrahams *et al.* investigated how $g(L)$ should evolve from Eq. (7.8) towards Eq. (7.9) by smoothly increasing L and by assuming that $g(L)$ is continuous [124]. Such an evolution should depend on the dimensionality of the system d . In one dimension, for instance, all states are known to be localized [122]. Thus in this case the only relevant limiting case is Eq. (7.9). In order to understand the behavior of $g(L)$ in a general case, they defined the *scaling function*

$$\beta = \frac{d \log g}{d \log L}, \quad (7.10)$$

and argued that β should be a continuous and smooth function of the conductance g *only*. In

one and two dimensions, $\beta(g)$ is always negative. Therefore by increasing L , g decreases, β becomes more and more negative and hence the system tends towards strong localization as $L \rightarrow \infty$. In three dimensions, the critical conductance g_c for the localization transition is expected to occur for $\beta(g_c) = 0$. In the limiting case of large conductance, $g \gg g_c$, Ohm's law Eq. (7.8) applies and $\beta(g)$ tends to the asymptotic form:

$$\beta(g) = d - 2 \quad (g \gg g_c). \quad (7.11)$$

On the other hand for small conductance, $g \ll g_c$, the scale dependence of $\beta(g)$ is given by Eq. (7.9) and its asymptotic form is independent of the dimensionality:

$$\beta(g) = \log(g/g_c) \quad (g \ll g_c). \quad (7.12)$$

In between the asymptotic forms (7.11) and (7.12), $\beta(g)$ can be calculated using the diagrammatic perturbation theory [115]:

$$\beta(g) = (d - 2) - a/g, \quad (7.13)$$

where a/g is the weak localization correction. The dependence of the scaling function β on the conductance g can thus be inferred as shown in Fig. 7-1. By inspection of the curve corresponding to the three dimensional case in Fig. 7-1, one distinguishes two regimes:

i) $g > g_c$

In this regime β is positive which implies that g increases as the system size L increases, which is consistent with the ohmic, classical transport scenario. This also leads to a further increase in β and thus one moves upwards along the β curve towards the asymptotic form 7.11.

ii) $g < g_c$

In this case β is negative and by increasing L one moves towards the strongly localized regime, characterized by the asymptotic form 7.12.

Finally, it is important to stress that the assumption of the continuity of the scaling function $\beta(g)$ implies that the localization transition is second-order, in contrast to a localization scenario proposed by Mott, who claimed the existence of minimum conductivity and, consequently, that

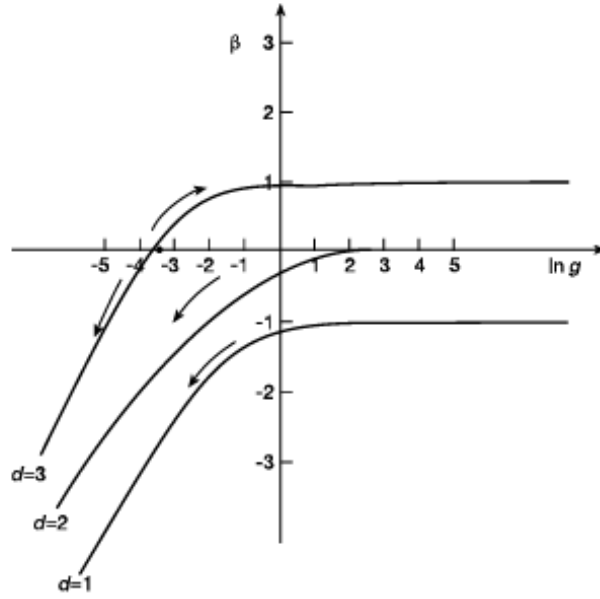


Figure 7-1: The qualitative behavior of the scaling function $\beta(g) = d \log g / d \log L$ in one, two and three dimensions.

the Anderson localization is a first-order phase transition [114].

7.3 Localization of light

John was among the first to realize that photons, like electrons, can be subject to localization effects, despite their different spin and dispersion law [125]. Moreover, photons do not mutually interact in contrast to electrons. In view of this fact, Anderson localization of electromagnetic waves could, at least in principle, be achieved more easily than in the electronic case since a metal-insulator transition is usually a result of a complicated interplay between disorder, associated with the Anderson transition, and electron correlations, associated with the Mott transition [114]. In this sense, electromagnetic localization could be regarded as a “pure” disorder-induced Anderson transition.

However, unlike electrons in a semiconductor, the number of photons is not always con-

served: they can be absorbed by matter. This fact complicates considerably the interpretation of experiments on electromagnetic localization. Indeed, the observation of an exponential scaling of transmission⁵, as expected in the localized regime, both for microwaves [126] and light [119][120][121] may not have definitively established electromagnetic localization since absorption could be responsible for this same effect. The same applies to the observation of the rounding of the backscattering cone [127], which may also be a manifestation of the onset of the localized regime, as suggested by recent theoretical predictions [128].

In view of these facts, the observation of electromagnetic wave localization is far from being a closed issue and alternative manners to detect it, that circumvent the difficulties related to absorption, are required. A recent and important progress in this sense was made by Chabanov, Stoytchev and Genack [129]. They have shown that the measurement of the variance of relative fluctuations of certain transmission quantities seems to provide an accurate criterion for electromagnetic localization, even in the presence of strong absorption. The main results of the second part of the thesis, to be presented in Chapter 9, also go in this same direction. We will investigate how localization effects manifest themselves in the decay rate statistics of optical open media and propose an alternative criterion for Anderson localization of light based on this investigation.

⁵See Eq. (7.9).

Résumé du chapitre 7 : Localisation d'ondes

Dans ce chapitre nous nous intéressons au phénomène de la localisation forte des ondes. Le concept de localisation forte a été introduit par P.W. Anderson en 1958 comme l'inhibition de la diffusion en raison des effets d'interférence entre les ondes qui subissent des diffusions multiples dans un milieu désordonné [10]. Anderson s'intéressait au transport électronique dans les semiconducteurs dopés mais, étant un phénomène essentiellement ondulatoire, la localisation forte peut se produire, du moins en principe, pour n'importe quel type d'onde. Pour la lumière, qui nous intéressera particulièrement dans la suite de la thèse, l'observation de la localisation forte [119] a été l'objet d'une intense controverse [120][121] et est toujours un sujet très discuté.

Les fonctions d'onde en régime localisé se caractérisent par une décroissance exponentielle :

$$|\psi(\mathbf{r})| \sim \exp(-|\mathbf{r} - \mathbf{r}_0|/\xi), \quad (7.14)$$

où ξ est la longueur de localisation qui mesure l'extension spatiale de la localisation. Comme un grand nombre d'expériences sont réalisées dans les milieux ouverts, il est important de considérer d'autres critères et définitions pour la localisation forte qui prennent en compte la fuite des ondes vers l'extérieur. Cette notion constitue la base du critère de Thouless pour la localisation que nous discuterons dans la suite.

Principaux critères pour la localisation

Le *critère de Thouless* [113] repose sur la comparaison entre deux échelles de temps : le temps de Thouless, t_T , et le temps de Heisenberg, t_H . Le temps de Thouless correspond au temps typique qu'une excitation nécessite pour traverser tout le système de taille L , $t_T \sim L^2/D$, où D est la constante de diffusion. Le temps de Heisenberg caractérise l'échelle de temps maximale du système, $t_H \sim 1/\Delta E$, où ΔE est le espacement moyen entre niveaux. Le critère de Thouless établit que la localisation se produit quand $t_T > t_H$, ce qui est incompatible avec le processus de diffusion pour lequel t_T doit être plus petit que t_H . En termes d'échelles d'énergie, le critère de Thouless pour la localisation s'écrit :

$$g \sim \rho(E)D(E)L^{d-2} < 1, \quad (7.15)$$

où g est la conductance adimensionnelle, d la dimension du système et $\rho(E)$ la densité d'états.

Le *critère de Ioffe-Regel* [114] pour la localisation dans les systèmes désordonnés est basé sur la comparaison entre la longueur d'onde λ et le libre parcours moyen ℓ . En $3D$, le critère de Ioffe-Regel établit que la localisation se produit quand λ est de l'ordre de ℓ :

$$k\ell < \text{constante} \approx 1, \quad (7.16)$$

où $k = 2\pi/\lambda$ est le nombre d'onde. Des calculs plus détaillés, basés sur l'analogie du puits de potentiel, montrent que la constante dans (7.16) vaut approximativement 0.84 [122].

Finalement, la *théorie d'échelle* pour la localisation [124] a pour but étudier le comportement de la conductance en fonction de la taille du système $g(L)$, en faisant l'hypothèse que $g(L)$ est continue et en supposant la forme de $g(L)$ connue pour les régimes localisé et diffus. Une des principales conclusions de la théorie d'échelle c'est que la localisation forte est une transition de phase de deuxième ordre, contrairement à ce que croyait Mott qui soutenait l'idée de l'existence d'une conductivité minimale [114].

Chapter 8

Decay rate statistics and the Anderson transition

This chapter is devoted to an overview of recent results concerning the influence of Anderson localization on decay rate statistics. Three regimes related to the Anderson transition will be discussed: the diffusive or extended regime, the localized regime and the critical point. As was anticipated in Chapter 6, the localized regime is beyond the scope of standard “Wigner-Dyson” Random Matrix Theory (RMT). The same applies for the critical point. Even for extended states, the application of the standard “Wigner-Dyson” RMT is restricted to the ballistic regime and limited by the existence of the so-called prelocalized states [130] in the diffusive regime. Alternative approaches to the standard “Wigner-Dyson” RMT should thus be employed to obtain decay rate statistics, such as diagonalizing of the Anderson Hamiltonian or using concepts of quantum chaos theory.

8.1 Localized regime

The first attempt to understand the behavior of the distribution of resonance widths $P(\Gamma)$ in the localized regime was carried out for quantum chaotic systems [131]. The phenomenon of localization is known to occur not only in disordered systems but also in quantum chaos. It is known that the presence of chaos [132] can be a substitute for disorder in quantum systems with a well-defined classical counterpart, a phenomenon referred to as *dynamical localization* [133]. In

systems exhibiting dynamical localization, chaos ‘mimics’ disorder and the “eigenstates” become exponentially localized, as in the Anderson scenario. The concept of dynamical localization was first developed for the kicked rotor model, where localization occurs in angular momentum space and the probability of finding high angular momenta is exponentially small at all times [133]. This explains the name dynamical localization. Strongly perturbed atomic [134] and molecular [135] systems are good candidates to study Anderson localization in quantum systems without the presence of disorder.

It was shown that in dynamically localized systems $P(\Gamma)$ should follow an inverse power law [131][136]:

$$P(\Gamma) \sim \frac{1}{\Gamma}. \quad (8.1)$$

This behavior was also recently reported for disordered systems, both in numerical calculations of the one-dimensional and quasi-one-dimensional Anderson model [137] and in analytical calculations [138], also in one-dimension.

Let us now try to understand qualitatively the result (8.1). As discussed in Chapter 7, the eigenstates of a closed, localized system are exponentially localized and hence their intensities at the boundaries are usually fairly small. This means that most of the eigenstates in the localized regime will be affected very weakly by the opening of the system. Therefore it is natural to treat localized resonances ϕ_n in an open system as a perturbation of the eigenstates Ψ_n of the closed system. Using Eq. (6.3), time-independent perturbation theory gives the first-order correction Δ_n to the (real) eigenvalues E_n of the (Hermitian) Hamiltonian $\mathcal{H}_{clo}$ of the closed system due to the opening of the system

$$\Delta_n = \mathcal{E}_n - E_n \sim -i \langle \Psi_n | W^\dagger W | \Psi_n \rangle. \quad (8.2)$$

In Eq. (8.2), $\mathcal{E}_n$ are the eigenvalues associated with the resonances ϕ_n of the total Hamiltonian $\mathcal{H}_{tot}$ of the open system. Since $\mathcal{H}_{tot}$ is non-Hermitian, its eigenvalues are complex $\mathcal{E}_n = E_n -$

$i\Gamma_n/2$ and it follows from Eq. (8.2) that the resonance widths Γ_n are given by

$$\begin{aligned}\Gamma_n &\sim \langle \Psi_n | W^\dagger W | \Psi_n \rangle \\ &= \sum_{m \in \text{boundary}} |\Psi_n(r_m)|^2 \approx |\Psi_n(r_n)|^2\end{aligned}\quad (8.3)$$

with $|\Psi_n(r_n)|^2$ the intensity of a localized eigenstate situated at a distance r_n from the sample boundary. As we know from Chapter 7 that $\Psi_n(r_n)$ is exponentially localized, the eigenstates intensities will be given by

$$|\Psi_n(r_n)|^2 \sim \exp(-2r_n/\xi) \exp(\sigma_n), \quad (8.4)$$

where we have introduced a fluctuating quantity σ_n to take into account possible intensity fluctuations that are expected to occur in the localized regime [129]. Using Eq. (8.4), Eq. (8.3) yields:

$$\Gamma_n \sim |\Psi_n(r_n)|^2 \sim \exp(-2r_n/\xi) \exp(\sigma_n). \quad (8.5)$$

Assuming that the resonances are uniformly distributed in space, it follows that the (integrated) probability of finding a resonance width Γ smaller than Γ_n , $P_{int}(\Gamma < \Gamma_n)$, is equal to the probability of finding a resonance situated at a distance r from the boundaries larger than r_n , $P(r > r_n)$, i.e., $P_{int}(\Gamma < \Gamma_n) = P(r > r_n)$. Since $P(r > r_n) \propto \mu_d(L - r_n) / \mu_d(L)$ with μ_d the d -dimensional volume and L the linear size of the system, we conclude that the probability density is

$$\begin{aligned}P(\Gamma_n) &= \frac{dr_n}{d\Gamma_n} \frac{d}{dr_n} [P(r > r_n)] \\ &\propto -\frac{\xi}{\Gamma_n} \frac{d}{dr_n} \left[\frac{\mu_d(L - r_n)}{\mu_d(L)} \right],\end{aligned}\quad (8.6)$$

which yields the power law decay (8.1) in the localized regime. Notice that the purely geometrical factor $d/dr_n [\mu_d(L - r_n) / \mu_d(L)]$ depends on the dimensionality of the system but does not affect the exponent in Γ . Notice also that the factor σ_n introduced in Eq. (8.4) to describe fluctuations does not contribute to $P(\Gamma_n)$ in view of the derivative $dr_n/d\Gamma_n$ in Eq.

(8.6). In the next chapter, it will be shown by means of extensive numerical simulations that the algebraic decay (8.1) describes very well the distribution of resonance widths for one-, two- and three-dimensional multiple light scattering systems in the localized regime.

However, the power-law decay (8.1) is not expected to be valid for all values of Γ , but rather restricted to some range of intermediate Γ values [137][138]. For the one-dimensional Anderson model (linear size L) it was shown that the range of Γ values for which the power law decay applies depends on the localization length ξ : $\exp(-L/\xi) < \Gamma/\Delta \ll 1$, with Δ the mean level spacing [137][138]. The quantity $\exp(-L/\xi)$ typically corresponds to the average value of Γ in the localized regime. Thus the region of algebraic decay in $P(\Gamma)$ tends to be broader as more and more states become localized.

For very small Γ , $\Gamma/\Delta < \exp(-L/\xi)$, $P(\Gamma)$ exhibits a different behavior. Sommers, Fyodorov and Titov have argued that, in the context of quantum chaotic systems, $P(\Gamma)$ should follow a log-normal distribution [139]. This behavior was reported later in numerical calculations for the two-dimensional Anderson model [94]. In order to understand this behavior qualitatively, one should recall that as one approaches the region $\Gamma/\Delta < \exp(-L/\xi)$ more and more resonances become localized (extremely small leakage), with a spatial distribution that is no longer expected to be uniform, but rather with resonances concentrated deep inside the sample, very far from the boundaries. Thus the argument leading to the power law decay of $P(\Gamma)$ [Eq. (8.1)] does not apply here. On the other hand, for $\Gamma/\Delta < \exp(-L/\xi)$ fluctuations in the resonances intensities become increasingly important and the factor $\exp(\sigma_n)$ dominates in Eq. (8.5). If one assumes a Gaussian distribution for σ_n , then in view of Eq. (8.5) $\ln \Gamma_n$ will also follow a Gaussian distribution and, as a result, $P(\Gamma_n)$ will be distributed log-normally. Notice however that this contribution is only due to resonances situated very far from the boundaries (typically a fraction ξ/L of the total number of resonances) and it is expected to become negligible for large r_n , i.e., for $\Gamma/\Delta > \exp(-L/\xi)$.

On the other hand, for very large Γ the resonances are very short-living and strongly coupled to the continuum. In this statistically less populated region, $P(\Gamma)$ decays much faster than algebraically [137].

8.2 Diffusive regime

For disordered mesoscopic systems, the standard “Wigner-Dyson” RMT applies only if wave transport is ballistic, i.e., when the disorder is insufficient to generate multiple scattering, which occurs for a mean free path ℓ much larger than the system size L , $\ell \gg L$. In this case, the distribution of resonance widths $P(\Gamma)$ was obtained using standard RMT techniques [97]. As the disorder increases, but not too much to reach the Anderson transition, multiple scattering sets in, the system becomes diffusive and deviations from RMT become increasingly apparent. These deviations are even more evident from the appearance of the so-called *prelocalized* or *anomalously localized states* (for a review, see Ref. [130]). These states exhibit strong localization features, such as an exponential spatial decay as in (7.2), although wave transport is, on average, still diffusive. For this reason, prelocalized states are considered as precursors of Anderson localization. They are typically situated far from the system boundaries and somehow hardly ‘feel’ the opening of the sample. In addition, they are rare compared to the typical extended states in the diffusive regime [130]. Despite being rare, prelocalized states are able to influence the behavior of different physical quantities in the diffusive regime. For instance, they have been proposed as the origin of the long tails in the probability distribution of the conductance and other physical observables in electronic transport, in strong contrast with the RMT predictions [140]. In optical disordered systems, prelocalized modes have recently been experimentally reported for a diffusive sample [141]. They are crucial for the understanding of the lasing threshold of random lasers which is determined by the mode with the smallest decay rate¹. The observation of laser action in disordered media far above the localization threshold has been interpreted as a manifestation of the existence of prelocalized states [141].

The mean value of Γ in the diffusive regime is typically the Thouless energy, $\Gamma_T \sim D/L^2$, inversely proportional to the time that an excitation needs to traverse the whole sample of size L (with D the Boltzmann diffusion constant). The Thouless energy marks two distinct regimes in the distribution of resonance widths, small Γ ($\Gamma \lesssim \Gamma_T$) and large ($\Gamma \gtrsim \Gamma_T$), for which $P(\Gamma)$ is expected to exhibit different behaviors. These regimes will be discussed bellow, following the arguments of Ref. [142]. As far as we know, there is no theory to describe the behavior

¹See section 6.1.2.

of $P(\Gamma)$ in the diffusive regime and Ref. [142] presents only heuristic arguments supported by numerical simulations. Apart from Ref. [142], where $P(\Gamma)$ was investigated for two-dimensional systems, the only work treating $P(\Gamma)$ in the diffusive regime seems to be Ref. [143], where the quasi-one-dimensional case is considered.

In the small Γ region ($\Gamma \lesssim \Gamma_T$) prelocalized modes are expected to manifest themselves in the distribution $P(\Gamma)$ of resonance widths [142]. They correspond to very sharp resonances with $\Gamma \ll D/L^2$. Since they are typically located at the bulk of the sample, they are affected very weakly by the opening of the system. These anomalously localized states in the diffusive regime are supposed to behave in the same way of ordinary localized states. Hence it is reasonable to apply the same arguments of the previous section for very small Γ 's in the localized regime and thus to expect a log-normal distribution for $P(\Gamma)$. Numerical simulations for the two-dimensional kicked rotor model confirm indeed that $P(\Gamma)$ follow a log-normal distribution in the diffusive regime for the region $\Gamma \lesssim \Gamma_T$ [142]. However, a more precise analysis indicates that a factor C_β should be included in the exponent of the distribution to take into account time reversal symmetry [144][142]:

$$P(\Gamma \lesssim \Gamma_T) \sim \exp \left[-C_\beta (\ln \Gamma)^2 \right]. \quad (8.7)$$

For the large Γ region ($\Gamma \gtrsim \Gamma_T$) in the diffusive regime the vast majority of modes are expected to be spread more or less uniformly throughout the entire medium, in contrast to localized or to prelocalized modes. Therefore, one can try to associate the lifetime of the mode with the typical time $t_R \sim 1/\Gamma_R \sim R^2/D$ that an excitation requires to reach the boundaries, when it is initially a distance R away. Notice that this classical picture is valid provided t_R do not exceed the time an excitation needs to traverse the entire sample $t_T \sim 1/\Gamma_T \sim L^2/D$, i.e., provided $\Gamma \gtrsim \Gamma_T$. Then the relative number of resonances that need a time $t < t_R$ to reach the boundaries (i.e., resonances with $\Gamma > \Gamma_R$) is

$$P_{int}(\Gamma_R) = \int_{\Gamma_R}^{\infty} P(\Gamma) d\Gamma \sim \frac{A(\Gamma_R)}{L^2}, \quad (8.8)$$

with $A(\Gamma_R)$ the area occupied for resonances that live less long than t_R . Equation (8.8) can be

written as

$$P_{int}(\Gamma_R) \sim \frac{\mu_d(L) - \mu_d(L - 2R)}{\mu_d(L)}, \quad (8.9)$$

with $\mu_d(L)$ the d -dimensional volume of size L . In three dimensions, $P_{int}(\Gamma_R)$ is

$$P_{int}(\Gamma_R) \sim \left(\frac{\Gamma_T}{\Gamma_R}\right)^{1/2} - 2\frac{\Gamma_T}{\Gamma_R} + \frac{4}{3}\left(\frac{\Gamma_T}{\Gamma_R}\right)^{3/2}. \quad (8.10)$$

For $\Gamma_R > \Gamma_T$ the first term in (8.10) dominates and the distribution of resonance widths is

$$P(\Gamma \gtrsim \Gamma_T) = -\frac{dP_{int}(\Gamma_R)}{d\Gamma_R}\bigg|_{\Gamma_R=\Gamma} \sim \sqrt{\frac{D}{L^2}} \frac{1}{\Gamma^{3/2}}. \quad (8.11)$$

A straightforward application of the above arguments shows that Eq. (8.11) should also be valid in two-dimensions for $\Gamma \gtrsim \Gamma_T$. Indeed, the power law behavior $P(\Gamma) \sim \Gamma^{-3/2}$ has been recently reported in numerical simulations for the two-dimensional kicked rotor model in the diffusive regime [142].

8.3 Critical point

In three dimensions, the region around the critical point of the Anderson transition is very particular, characterized by strong fluctuations on all scales. Much is known about the properties of eigenstates and eigenvalues of closed systems at the transition. In particular, it was shown that eigenstates exhibit multifractal behavior and that the level spacing distribution exactly at the transition is a crossover from the Poisson distribution, associated with the localized regime, to the Wigner distribution, associated with the diffusive regime [145].

However, much less is known about the statistical properties of open systems at critical conditions. For the distribution of resonance widths $P(\Gamma)$, in particular, the only existent work is due to Kottos and Weiss [146]. They have argued, using heuristic arguments, that $P(\Gamma)$ should follow an universal power law decay at the transition, but with powers different from

the ones for the localized and the diffusive regimes:

$$P(\Gamma) \sim \Gamma^{-(1+1/d)}, \quad (8.12)$$

with the d dimensionality of the system. In (8.12) the values of Γ have been normalized by the mean level spacing Δ . This prediction was confirmed by a numerical calculation using the three-dimensional Anderson model.

Résumé du chapitre 8 : Statistique des largeurs de résonance et la transition d'Anderson

Ce chapitre est consacré à une révision des principaux résultats concernant la statistique des largeurs de résonance $P(\Gamma)$ dans les situations associées à la transition d'Anderson : le régime diffus, le régime localisé et le point critique.

Régime localisé

Il a été montré pour les systèmes dynamiquement localisés que $P(\Gamma)$ suit une loi de puissance du type [131][136] :

$$P(\Gamma) \sim \frac{1}{\Gamma}. \quad (8.13)$$

Des calculs analytiques [138] et des simulations numériques [137] pour le modèle d'Anderson montrent que ce comportement est également valable pour les systèmes désordonnés uni- et quasi-unidimensionnels en régime localisé. Nous avons montré que le résultat (8.13) est *universel*, parce qu'il est indépendant de la dimension du système.

Néanmoins, la décroissance algébrique (8.13) n'est pas valable pour toutes les valeurs de Γ . Pour les Γ petits, $\Gamma/\Delta < \exp(-L/\xi)$ (où Δ est l'espacement entre niveaux et ξ la longueur de localisation), $P(\Gamma)$ suit une distribution log-normale [139]. Le cas $\Gamma/\Delta < \exp(-L/\xi)$ correspond typiquement à la valeur moyenne de Γ en régime localisé. Pour les Γ très grands ($\Gamma/\Delta \gg$

$\exp(-L/\xi)$, (8.13) ne s'applique pas non plus et $P(\Gamma)$ décroît plus rapidement qu'une loi de puissance [137].

Régime diffus

En régime diffus, la valeur moyenne de Γ est typiquement l'énergie de Thouless $\Gamma_T \sim D/L^2$. Pour les Γ petits ($\Gamma \lesssim \Gamma_T$), les états prélocalisés jouent un rôle important dans le calcul de $P(\Gamma)$. Ces états possèdent des caractéristiques similaires à celles des états localisés malgré le fait que le système se trouve, en moyenne, dans le régime diffus [130]. Bien que rares parmi les états étendus, ils influencent la distribution $P(\Gamma)$ qui suit, pour $\Gamma \lesssim \Gamma_T$, une distribution log-normale [144][142]:

$$P(\Gamma \lesssim \Gamma_T) \sim \exp \left[-C_\beta (\ln \Gamma)^2 \right], \quad (8.14)$$

où C_β est un facteur qui prend en compte la symétrie de renversement temporel.

Pour $\Gamma \gtrsim \Gamma_T$, des calculs numériques récents montrent que $P(\Gamma)$ suit une loi de puissance [142]:

$$P(\Gamma) \sim \Gamma^{-3/2}. \quad (8.15)$$

Point critique

Au point critique de la transition d'Anderson en 3D, Kottos et Weiss ont proposé, en utilisant des arguments heuristiques et des simulations numériques, que $P(\Gamma)$ suit une loi de puissance [146]:

$$P(\Gamma) \sim \Gamma^{-(1+1/d)}, \quad (8.16)$$

où d est la dimension du système.

Chapter 9

Probing Anderson localization of light via decay rate statistics

Probing Anderson localization of light via decay rate statistics

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Abstract

We have studied the distribution of resonant widths $P(\Gamma)$ in one-, two- and three dimensional multiple light scattering systems. $P(\Gamma)$ should follow a universal power law $P(\Gamma) \sim \Gamma^{-1}$ in the localized regime as confirmed by extensive numerical calculations. This behavior can be interpreted as an unambiguous signature of exponential Anderson localization of light in open systems.

The research on Anderson localization of light has been of great interest [6] since it was originally proposed as the optical counterpart of electronic localization [2]. Localization, as proposed by Anderson, is defined as an inhibition of wave diffusion in infinite disordered media due to interference of multiple scattered waves [3]. A much stronger definition is that the eigenfunctions in an infinite disordered medium are characterized by an exponential decay in space, $|\psi(\mathbf{r})| \sim \exp(-|\mathbf{r} - \mathbf{r}'|/\xi)$, where ξ is the localization length. In finite, *open* media, waves can “leak” through the sample boundaries. Anderson localization must thus relate to manifestations of leakage in observables quantities. For optical systems, they are typically the emerging intensity, the total transmission or the coherent backscattering cone. The observation of an exponential scaling of transmission [4, 5], as well as the rounding of the backscattering cone [6], may not have definitively established localization since absorption could be responsible for these same effects.

There are several criteria to determine the onset of the localized regime. The Ioffe-Regel criterion states that, in three dimensions (3D), localization occurs for $k\ell \sim 1$ (with k the light wavenumber inside the medium and ℓ the mean free path). Another approach observes electromagnetic localization from the variance of fluctuations of transmission, even in the presence of absorption [7]. In open systems, the “eigenstates” are resonances with a finite energy width Γ (or, equivalently, with a finite lifetime $t \sim 1/\Gamma$) due to leakage. The Thouless criterion asserts that localization can be said to occur when the typical time that an excitation needs to propagate through the entire system of size R , $t_T \sim 1/\Gamma_T \sim R^2/D$ (Thouless time), exceeds the maximal time scale of the system, $t_H \sim 1/\Gamma_H \sim 1/\Delta E$ (Heisenberg time) [8]. Here D is the diffusion constant and ΔE the mean level spacing.

This Thouless criterion applies to the *average* leakage width. Hence it is reasonable to assume that the *statistical properties* of resonance widths are strongly affected by localization. The aim of the present paper is to investigate how localization manifests itself in the distribution of resonance widths $P(\Gamma)$ in multiple light scattering in open systems. We will show that $P(\Gamma)$ exhibits the universal power law $P(\Gamma) \sim \Gamma^{-1}$ in 1D, 2D and 3D *optical* disordered systems, thereby generalizing recent theoretical [9] and numerical [10] studies in 1D models of mesoscopic transport. We assert that the algebraic decay $P(\Gamma) \sim \Gamma^{-1}$ represents a universal property of Anderson localization of light in open systems of any dimension.

Although the statistical properties of resonance widths in open systems have been extensively studied over the last years, in particular for chaotic/ballistic systems [11, 12, 13], their behavior for disordered systems exhibiting localization has received considerably less attention. As argued by Casati *et al.*, $P(\Gamma)$ should follow a power law $P(\Gamma) \sim \Gamma^{-1}$ in localized, classically chaotic systems [14]. $P(\Gamma)$ was analytically obtained for 1D disordered systems, showing a slightly different power law $P(\Gamma) \sim \Gamma^{-1.25}$ [9]. This prediction was corroborated later by numerical calculations in 1D and quasi-1D tight binding models [10]. The $P(\Gamma) \sim \Gamma^{-1}$ behavior was also reported in 1D and 3D strongly driven atomic Rydberg states in the context of dynamical localization [15]. Exactly at the Anderson transition [16] and in the diffusive regime [17], $P(\Gamma)$ was shown to follow a power law with a power different from -1 . Concerning the study of $P(\Gamma)$ for optical systems, the only work on the subject is, to the best of our knowledge, due to Patra [18], who mainly focused on the small Γ regime and its application to random lasers. For small Γ (i.e. for $\Gamma \lesssim \langle \Gamma \rangle$), it is known that $P(\Gamma)$ is different from a power law, for both the diffusive [12, 18] and the localized [13, 18] regimes. It should be emphasized that the power law decay of $P(\Gamma)$ is expected to occur only for $\Gamma \gtrsim \langle \Gamma \rangle$, with typically $\langle \Gamma \rangle \sim \Gamma_T$ in the diffusive regime and $\langle \Gamma \rangle \sim \exp(-R/\xi)$ in the localized regime. However, for very large Γ ($\Gamma \gg \langle \Gamma \rangle$) the resonances are strongly coupled to the continuum and $P(\Gamma)$ decays faster than algebraically, both in the diffusive [17] and in the localized [9, 10] regimes.

We will present a simple physical argument, inspired by Refs. [14, 9, 10], to explain the *universal* $P(\Gamma) \sim \Gamma^{-1}$ behavior for the localized regime, i.e., independent of the dimensionality of the system. Due to the opening of the system, exponentially localized eigenstates of the corresponding closed system (linear size R) acquire a finite frequency width Γ' , $\Gamma' \sim e^{-2r'/\xi}$, with r' the distance to the boundaries. Near the system boundaries, the leakage is strong and the resonances are broad compared to Γ_T . On the other hand, far from the boundaries the leakage is small and the typical Γ in this region is much smaller than Γ_T . Assuming that the resonances are - like the scatterers - uniformly distributed in space, it follows that the (integrated) probability of finding a resonance width Γ smaller than Γ' , $P_{int}(\Gamma < \Gamma')$, is equal to the probability of finding a resonance situated at a distance r from the boundaries larger than r' , $P(r > r')$, i.e., $P_{int}(\Gamma < \Gamma') = P(r > r')$. Since $P(r > r') \propto \mu_d(R - r') / \mu_d(R)$ with

μ_d the d-dimensional volume, we conclude that the probability density is

$$\begin{aligned} P(\Gamma') &= \frac{dr'}{d\Gamma'} \frac{d}{dr'} [P(r > r')] \\ &\propto -\frac{\xi}{\Gamma'} \frac{d}{dr'} \left[\frac{\mu_d(R - r')}{\mu_d(R)} \right]. \end{aligned} \quad (9.1)$$

The purely geometrical factor $d/dr' [\mu_d(R - r') / \mu_d(R)]$ depends on the dimensionality of the system but does not affect the exponent in Γ .

To test the validity of (9.1) for Anderson localization of light, we will consider scalar wave propagation in disordered media using the method introduced by Rusek and Orłowski [19, 20]. This approach is based on the analysis of the spectrum of the Green matrix, that describes light scattering from randomly distributed pointlike dipoles (i.e., particles much smaller than the wavelength of light). For an incident plane wave $\psi_0(\mathbf{r})$ in a system of N identical dipoles with scattering matrix t , the field acting in the dipole at $\mathbf{r}_i$ is given by [19, 20]:

$$\psi(\mathbf{r}_i) = \psi_0(\mathbf{r}_i) + t \sum_{j \neq i}^N G(\mathbf{r}_{ij}) \psi(\mathbf{r}_j). \quad (9.2)$$

The complex-valued $N \times N$ matrix $G(\mathbf{r}_{ij})$ describes light propagation of the wave scattered by the dipole at $\mathbf{r}_i$ to the dipole at $\mathbf{r}_j$. Since the eigenvalues λ_M of $\mathbf{M} \equiv \mathbf{I} - t\mathbf{G}$ and λ_G of $\mathbf{G}$ are related by $\lambda_M = 1 - t\lambda_G$, and t depends on frequency ω via the scattering phase shift $\delta(\omega)$ [21], an eigenvalue λ_G with $\text{Re}\lambda_G = -1$ will facilitate an appropriate choice for $\delta(\omega)$ such that $\lambda_M = 0$. This would correspond to a genuinely localized state somewhere inside the random medium [19]. Assuming a Breit-Wigner model for the scatterers (with one sharp resonance of width Γ_0 at the position ω_0), for which $\delta(\omega)$ has a simple form, it is possible to obtain, in a good approximation, the resonance widths Γ via λ_G , $\Gamma/\Gamma_0 \simeq 1 + \text{Re}\lambda_G$ [20]. We will numerically diagonalize $\mathbf{G}$ in 1D, 2D and 3D and calculate the distribution of resonance widths $P(\Gamma)$ using the above approximation.

It is interesting to compare the typical values of the resonance widths Γ to the Thouless frequency Γ_T . To estimate Γ_T let us recall that $\Gamma_T = 1/t_T = 2dD_B/R^2$, where D_B is the Boltzmann diffusion constant and d the dimension of the system. The Boltzmann diffusion constant is given by $D_B = v_E \ell^*/d$, where ℓ^* is the transport mean free path (which is, for

point scatterers, equal to ℓ) and v_E the energy transport velocity, $v_E \approx c_0/[1 + \tau_{dwell}/\tau_{mf}]$ [49], with $\tau_{dwell} = 1/\Gamma_0$ the dwell time in a single scattering and $\tau_{mf} = \ell/c_0$ the mean free time. The mean free path is given by $\ell = 1/n\sigma_d$, with n the density of scatterers and σ_d the $(d-1)$ dimensional cross-section of a single point scatterer. Applying these considerations, Γ_T can be written as $\Gamma_T/\Gamma_0 \sim 2(\ell/R)^2$. In what follows, we will always compare the values of Γ to Γ_T .

In Fig. (9-1), $P(\Gamma)$ is calculated for 1D systems composed of 100 randomly distributed scatterers in a linear segment for two different values of the uniform optical density ρ : $\rho = 1$ and $\rho = 10$ scatterers per wavelength. In 1D, all eigenstates are known to be exponentially localized even for weak disorder and ξ is of the order of the mean free path ℓ . $P(\Gamma)$ is seen to exhibit a power law with an exponent very close to -1 , in good agreement with (9.1). In addition, the exponent does not change by increasing ρ , i.e., by decreasing ξ . This demonstrates that the algebraic decay $P(\Gamma) \sim \Gamma^{-1}$ in the localized regime is valid not only for 1D models of mesoscopic transport [9, 10], but also for our model of wave propagation in disordered media. At large Γ , $P(\Gamma)$ decays faster than algebraically. This can be explained by the fact that this region is dominated by short living resonances, typically close to the boundaries, for which the prediction (9.1) breaks down. To compare the values of Γ to the Thouless frequency Γ_T , let us recall that the (dimensionless) cross-section of a point scatterer in 1D is simply the reflection coefficient. This implies that on resonance, $\Gamma_T/\Gamma_0 \sim 2(\ell/R)^2 = 2/N^2$ with N the number of scatterers. For $N = 100$ as in Fig. (9-1), we have $\Gamma_T/\Gamma_0 \simeq 0.0002$, showing that the values of Γ in Fig. (9-1) are far above the Thouless frequency Γ_T . The Thouless frequency Γ_T does not represent the appropriate characteristic decay rate for 1D systems since diffusion never occurs.

Fig. (9-2) shows $P(\Gamma)$ for 2D systems containing $N = 2500$ scatterers randomly distributed in a $R \times R$ square for $\rho = 1$ and $\rho = 10$ scatterers per wavelength squared. In 2D, in principle all eigenstates are exponentially localized but the localization length ξ may be macroscopically large for low disorder according to [23]:

$$\xi \simeq \ell \exp(\pi k_e \ell / 2), \quad (9.3)$$

with k_e the effective wavenumber, which takes into account renormalized diffusion. Localization is expected to occur when ξ is smaller than the system size R . The Γ^{-1} decay of $P(\Gamma)$ in

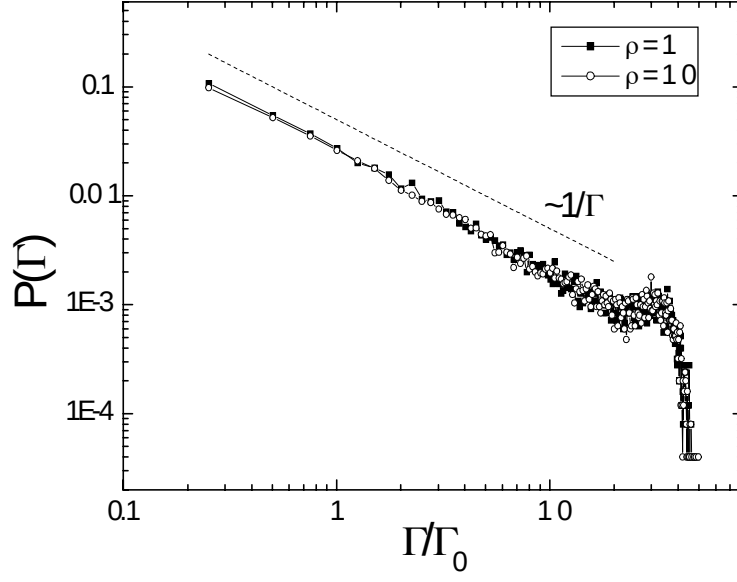


Figure 9-1: The normalized distribution of resonance widths $P(\Gamma)$ calculated for 1000 different configurations of 100 point scatterers randomly distributed in a 1D segment with two different values of the uniform optical density ρ , $\rho = 1$ (full squares) and $\rho = 10$ (open circles) scatterers per wavelength. The dashed line corresponds to the prediction $P(\Gamma) \sim \Gamma^{-1}$ for the localized regime and the solid lines are just to guide the eyes. The values of Γ are normalized by the resonance width of a single dipole Γ_0 . The value of the Thouless frequency for resonant scatterers is $\Gamma_T/\Gamma_0 = 2/N^2 \simeq 0.0002$.

Fig. (9-2) is clearly visible for both values of ρ used, with an exponent very close to -1 , in excellent agreement with (9.1). Notice that the range of the power law broadens as ρ increases. Increasing ρ means decreasing ℓ and, according to Eq. (9.3), a rapidly decreasing ξ . The range of the algebraic decay $P(\Gamma) \sim \Gamma^{-1}$ is expected to be broader as more and more states become localized. Such a behavior was also reported in numerical calculations within the Anderson model [10]. For large Γ , $P(\Gamma)$ decays again faster than algebraically as in the 1D case. To confirm that the system is indeed in the localized regime, let us estimate the ratio ξ/R from Eq. (9.3). Since $\ell/R = \pi/(2\sqrt{N\rho})$, the system can be said to be localized ($\xi/R < 1$) when $k_e\ell \approx 2$ for $N = 2500$ and for both values of ρ ($\rho = 1$ and $\rho = 10$) used in Fig. (9-2). This value of $k_e\ell$ is not too far from the “bare” estimates for the value of $k\ell = \pi^2/\rho$ in Fig. (9-2): $k\ell \approx 10$ for $\rho = 1$ and $k\ell \approx 1$ for $\rho = 10$, where we use the vacuum wavenumber $k = 2\pi/\lambda$ and not the effective wavenumber k_e . We conclude that the localized scenario is valid. Consequently, the Thouless frequency Γ_T is not the appropriate characteristic decay rate in this case, as in 1D. For resonant scatterers in 2D, the ratio of Γ_T to Γ_0 is $\Gamma_T/\Gamma_0 \sim 2(\ell/R)^2 = \pi^2/2N\rho$. For the values used in Fig. (9-2), we have $\Gamma_T/\Gamma_0 \simeq 0.002$ and $\Gamma_T/\Gamma_0 \simeq 0.0002$, corresponding to $\rho = 1$ and $\rho = 10$, respectively.

In Fig. (9-3) the 3D case is considered, where $P(\Gamma)$ is calculated for systems composed by 1000 point scatterers randomly distributed in a sphere (radius R) for $\rho = 1$, $\rho = 10$, $\rho = 30$ and $\rho = 60$ scatterers per wavelength cubed. In 3D, the system is expected to undergo, upon varying the degree of disorder, a transition from extended states to localized states. It is therefore interesting to investigate if and how this transition manifests itself in $P(\Gamma)$. As in the 2D case, we notice that, as ρ increases, the range of the algebraic decay $P(\Gamma) \sim \Gamma^{-\alpha}$ increases. We also remark that, as ρ increases, the *associated exponents tend more and more to the value* -1 . The exponents, obtained by a linear fit in the range where the power law is present, are $\alpha \approx 0.76$ for $\rho = 1$, $\alpha \approx 0.83$ for $\rho = 10$, $\alpha \approx 0.95$ for $\rho = 30$ and $\alpha \approx 1.1$ for $\rho = 60$. This suggests, according to (9.1), the onset of the localized regime for higher ρ . In fact, the Ioffe-Regel criterion for localization ($k\ell < 1$) is estimated to be satisfied for $\rho > 2\pi^2 \approx 20$ for scatterers at resonance. This condition is fulfilled for $\rho = 30$ and $\rho = 60$, for which α is very close to 1, showing that the system with these densities are indeed in the localized regime and confirming that the power law $P(\Gamma) \sim \Gamma^{-1}$ can be considered a genuine signature of Anderson

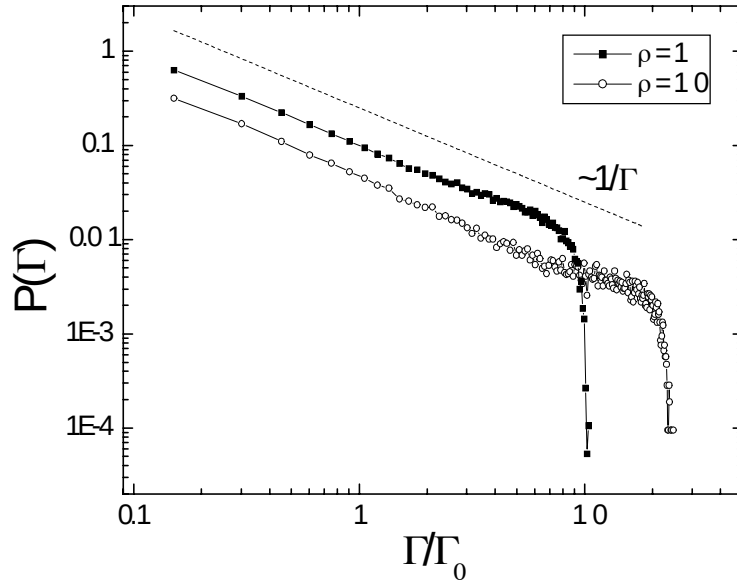


Figure 9-2: $P(\Gamma)$ calculated for up to 50 configurations of 2500 scatterers randomly distributed in a square for $\rho = 1$ (full squares) and $\rho = 10$ (open circles) scatterers per wavelength squared. The normalization of Γ , as well as the significance of the solid and dashed lines, is the same as in Fig. (9-1). The value of the Thouless frequency for resonant scatterers is $\Gamma_T/\Gamma_0 \simeq 0.002$ and $\Gamma_T/\Gamma_0 \simeq 0.0002$ for $\rho = 1$ and $\rho = 10$, respectively.

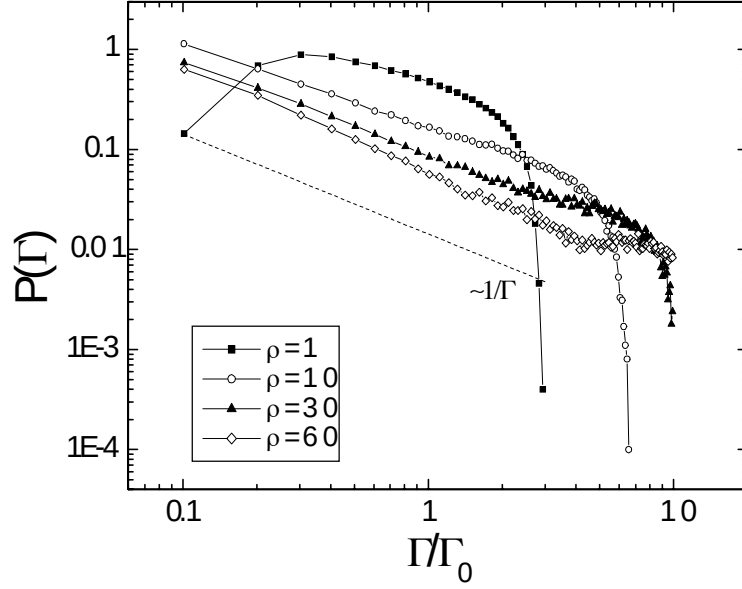


Figure 9-3: $P(\Gamma)$ calculated for 100 configurations of 1000 scatterers randomly distributed in a sphere for $\rho = 1$ (full squares), $\rho = 10$ (open circles), $\rho = 30$ (full triangles) and $\rho = 60$ (open diamonds) scatterers per wavelength cubed. The normalization of Γ , as well as the significance of the solid and dashed lines, is the same of Fig. (9-1).

localization of light. We anticipate that for $N, R \rightarrow \infty$ at constant ρ , the transition from the localized regime ($\alpha = 1$) to the diffusive regime will become even more evident. Once again, note that $P(\Gamma)$ decays faster than a power law for very large Γ .

In 3D, the Thouless frequency Γ_T is the real characteristic internal decay rate of system since here there is a real diffusive regime, in contrast to the 1D and 2D cases. For this reason, we exhibit in Fig. (9-4) $P(\Gamma)$ for the same optical densities of Fig. (9-3) but now with Γ normalized to the Thouless frequency Γ_T , with Γ_T given by $\Gamma_T/\Gamma_0 = 2(\ell/R)^2 = 2(4\pi/3)^{2/3} \times [\pi/(N^{1/3}\rho^{2/3})]^2$. For low ρ ($\rho = 1$ and $\rho = 10$), $P(\Gamma)$ is peaked near Γ_T , showing that the system is in the diffusive regime. Notice that there is a non-vanishing probability to find modes that live much longer than t_T even in the diffusive regime, the so-called “prelocalized” modes [24]. As ρ increases, we observe that $P(\Gamma)$ is no longer centered at Γ_T and that the probability to find a mode with resonance width smaller than Γ_T also increases. This means that, on average,

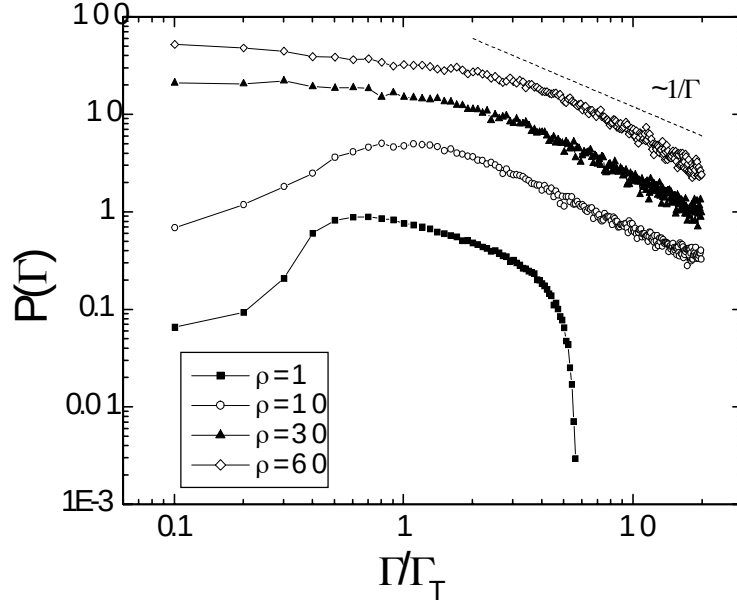


Figure 9-4: $P(\Gamma)$ as in Fig. (9-3), but now Γ is normalized by the Thouless frequency, Γ_T .

the modes live longer than t_T . At the same time, Fig. (9-3) shows that localization manifests itself in $P(\Gamma)$ not only via the broadening of the power law range but also via the fact that the associated exponents approach to -1 . We conclude again that the $P(\Gamma) \sim \Gamma^{-1}$ behavior is an unambiguous signature of Anderson localization of light in open media. It must be mentioned that the present 3D study may be relevant for recent multiple light scattering experiments in atomic media [25], for which modeling the scatterers by pointlike dipoles constitutes an excellent approximation, though with a varying density ρ .

In summary, we have studied the distribution of resonance widths $P(\Gamma)$ in 1D, 2D and 3D multiple light scattering systems composed of randomly distributed pointlike scalar dipoles. We have developed a simple physical argument, based on the exponential decay of localized eigenfunctions, to show that $P(\Gamma)$ should follow an universal power law $P(\Gamma) \sim \Gamma^{-1}$ decay in the localized regime. This prediction was confirmed by extensive numerical calculations and demonstrates that the $P(\Gamma) \sim \Gamma^{-1}$ behavior can be interpreted as an unambiguous signature of Anderson localization of light in open media.

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Résumé du chapitre 9 : Pour sonder la localisation d'Anderson de la lumière via la statistique de largeurs de résonance

Ce chapitre est consacré à l'étude de la distribution de largeurs de résonance $P(\Gamma)$ en systèmes optiques désordonnés ouverts en 1D, 2D et 3D.

En utilisant des arguments physique simples, basées sur la décroissance exponentielle des fonctions d'onde $|\psi(\mathbf{r})| \sim \exp(-|\mathbf{r} - \mathbf{r}_0|/\xi)$, nous avons montré que $P(\Gamma)$ suit une *loi de puissance universelle* (indépendant de la dimension du système) en régime localisé :

$$P(\Gamma) \sim \frac{1}{\Gamma}. \quad (9.4)$$

Ce résultat généralise ainsi des études numériques [137] et analytiques [138] récentes qui montrent que $P(\Gamma)$ suit un comportement du type (9.4) en régime localisé dans les systèmes de basse dimensionalité (1D et quasi 1D).

Nous avons confirmé le comportement (9.4) pour les systèmes optiques désordonnés en régime localisé en réalisant des simulations numériques intensives. Nous avons utilisé le modèle des diffuseurs ponctuels, introduit par Rusek et Orlowski [81] [82] pour décrire la diffusion multiple de la lumière dans les milieux désordonnés. Les simulations montrent que le comportement (9.4) peut être interprété comme une signature de la localisation forte dans les milieux optiques

désordonnés ouverts. Notre travail peut être pertinent pour l'étude de la localisation forte de la lumière dans les milieux atomiques [1], pour lesquels l'approximation des diffuseurs ponctuels est excellente.

Chapter 10

Decay rate statistics for vector waves: application to magneto-optics of open random media

An important extension of the work presented in Chapter 9 is to investigate the distribution of resonance widths $P(\Gamma)$ using a *vectorial* treatment of electromagnetic wave propagation and scattering. Such a treatment would provide a more realistic model to describe light propagation in disordered media, since it includes the polarization of the wave field. The use of a vectorial scenario for wave propagation could also shed some more light on the interplay between the existence of prelocalized states and the threshold of random lasers in the diffusive regime, which remains unclear so far. As already discussed, the threshold of a random laser is determined by the mode with the smallest decay rate Γ . The observation of localized states in disordered optical media with gain far from the localization threshold [141] seems to indicate that prelocalized states, with very small Γ , play a crucial role in the lasing instability of random media. In view of this experimental fact, it would be interesting to have a deeper understanding of the behavior of $P(\Gamma)$ for small Γ in diffusive optical systems. At the time of writing, the only work that analyzes $P(\Gamma)$ for small Γ in the diffusive regime for disordered systems seems to be Ref. [94], where the analysis is restricted to a scalar treatment of wave propagation for the two-dimensional Anderson model.

The use of a vectorial scenario for wave propagation also offers the possibility to investigate if and how an external magnetic field affects strong localization of light. Magnetic fields are known to affect coherent backscattering, explained by the weak localization theory as an interference effect between direct and reverse scattering paths in wave propagation [90]. This is true for electronic [90] and optical systems, for which the influence of magnetic fields in coherent backscattering has been extensively investigated over the last years, both theoretically [54, 57, 59, 147] and experimentally [148, 149, 150]. However, the impact of magnetic fields on Anderson localization has received considerably less attention, particularly for optical systems. Probably one of the most important existing theoretical works concerning Anderson localization in magnetic fields is due to Pichard *et al.* [151]. Using an extended version of Random Matrix Theory (RMT), they have argued that the localization length ξ should be proportional to the symmetry index ν as $N \gg 1$, $\xi \approx \nu N \ell$ (with N the number of propagating channels and ℓ the mean free path). A time-reversal symmetry breaking magnetic field induces a transition from $\nu = 1$ to $\nu = 2$, and thus just a doubling of ξ . Nevertheless this result only applies to low dimensional systems (one- and quasi-one dimensions) since it has been derived using a RMT-based approach, as discussed in Chapter 6. In view of this context, an important question arises: what are the effects of external magnetic fields on Anderson localization in higher dimensional systems and, in particular, higher dimensional optical systems? This motivation was stimulated by the first experiments on multiple light scattering in cold atomic clouds [152, 153, 154], including the observation of the coherent backscattering effect in cold Rubidium [152] and Strontium [154]. Modelling two-level atoms by identical point-dipoles with one internal resonance constitutes a very good approximation. Also, it has recently been reported that cold atoms exhibit high magneto-optical effects [155], which constitutes an additional motivation for the work to be presented in this chapter.

With these motivations in mind, in the present chapter we will investigate $P(\Gamma)$ for three-dimensional disordered systems using the vectorial version of the point-dipole model introduced by Rusek and Orłowski [81, 82, 156, 157, 158]. We will focus on the study of $P(\Gamma)$ in the *diffusive regime* and its behavior in the presence of an external magnetic field. To accomplish this, in section 10.1 we will first describe how to modify the point-dipole model in order to include the magneto-optical activity inside the dipoles. It will be shown how the distribution of eigenvalues

of the scattering G matrices is modified by an external magnetic field. The calculation of $P(\Gamma)$ and the discussion of the results is postponed to section 10.2, where we will also make a connection to possible applications to random lasers. Finally, in section 10.3 we will summarize the results and discuss some perspectives for future work.

10.1 Scattering G matrices in a magnetic field

In the presence of an external magnetic field $\mathbf{B}$, the 3×3 T -matrix $\mathbf{t}(k)$ describes the vector wave scattering by a single magneto-optical dipole. It was analytically calculated by Van Tiggelen, Maynard and Nieuwenhuizen, and exhibits scattering resonances at frequencies ω_0 (linewidth γ) and $\omega_0 \pm V B c_0$ (with V the Verdet constant of the scatterer), corresponding to the Zeeman levels $m = 0$ and $m = \pm 1$, respectively [54]. The total $3N \times 3N$ T matrix of an assembly of N magneto-optical scatterers situated at the positions $\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N$ is given by:

$$\mathbf{T}_{\mathbf{k}, \mathbf{k}'} = \begin{pmatrix} e^{i\mathbf{k} \cdot \mathbf{r}_1} \\ \vdots \\ e^{i\mathbf{k} \cdot \mathbf{r}_N} \end{pmatrix}^* \mathbf{t} \cdot (\mathbf{U} - \mathbf{G} \cdot \mathbf{t})^{-1} \cdot \begin{pmatrix} e^{i\mathbf{k}' \cdot \mathbf{r}_1} \\ \vdots \\ e^{i\mathbf{k}' \cdot \mathbf{r}_N} \end{pmatrix}, \quad (10.1)$$

where $\mathbf{U}$ is the $3N \times 3N$ unit matrix, $\mathbf{k}$ and $\mathbf{k}'$ are, respectively, the incident and the scattered wave vectors, and in the far field $|\mathbf{k}'| = |\mathbf{k}| = k = \omega/c_0$. The elements of the $3N \times 3N$ $\mathbf{G}$ -matrix are equal to the Green functions calculated from the relative positions of the N dipoles [49]:

$$\mathbf{G}_{NM}(\omega) = \begin{cases} -\frac{\exp(ikr_{NM})}{4\pi r_{NM}} \left\{ [\mathbf{U} - \hat{\mathbf{r}}_{NM} \hat{\mathbf{r}}_{NM}] - \left(\frac{1}{ikr_{NM}} + \frac{1}{(kr_{NM})^2} \right) [\mathbf{U} - 3\hat{\mathbf{r}}_{NM} \hat{\mathbf{r}}_{NM}] \right\} & \text{for } N \neq M, \\ 0 & \text{for } N = M. \end{cases} \quad (10.2)$$

Equation (10.1) determines the electric field scattered by the magneto-optical dipoles. It requires the diagonalization of the $3N \times 3N$ scattering matrix:

$$\mathbf{M}(\omega) \equiv \mathbf{t}(\omega) \cdot [\mathbf{U} - \mathbf{G}(\omega) \cdot \mathbf{t}(\omega)]^{-1}. \quad (10.3)$$

The T -matrix describing one single scatterer $\mathbf{t}(\omega)$ can be expressed by a Born series [48]:

$$\mathbf{t}(\omega) = \mathbf{V}(\omega) + \mathbf{V}(\omega) \cdot \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) + \dots = \left(\frac{1}{\mathbf{V}(\omega)} - \mathbf{G}_0(\omega, \mathbf{r} = 0) \right)^{-1}, \quad (10.4)$$

where $\mathbf{V}(\omega)$ is the optical potential for a point scatterer and $\mathbf{G}_0(\omega, \mathbf{r} = 0)$ is the return Green's function, i.e., the dyadic 3×3 Green's function $\mathbf{G}_0(\omega, \mathbf{r})$ evaluated at $\mathbf{r} = 0$. The optical potential is, in contrast to the potential for quantum mechanical scattering, frequency-dependent. For a atomic dipole at the position $\mathbf{r}_i$ subject to an external magnetic field $\mathbf{B}$ it takes the form

$$\mathbf{V}(\omega, \mathbf{r}, \mathbf{B}) = - \left(\frac{\omega}{c_0} \right)^2 \boldsymbol{\alpha}(\mathbf{B}) \delta(\mathbf{r} - \mathbf{r}_i), \quad (10.5)$$

where $\boldsymbol{\alpha}(\mathbf{B})$ is the polarizability tensor of the scatterer. It has the dimension of a volume which represents the microscopic size of the dipoles and is typically of the order of a_0^3 (a_0 the Bohr radius), but can be much bigger when higher Rydberg states are involved in the transition. For a magneto-optical dipole the polarizability $\boldsymbol{\alpha}(\mathbf{B})$ is given by:

$$\frac{1}{\boldsymbol{\alpha}(\mathbf{B})} = \frac{\mathbf{U}}{\alpha_0} + \mathcal{L}(\mathbf{B}), \quad (10.6)$$

where α_0 is the ordinary polarizability of the scatterer [48] and $\mathcal{L}(\mathbf{B})$ is the magneto-optical correction as a result of the Zeeman splitting. We shall restrict ourselves to linear orders in $\mathbf{B}$, in which case this correction is given by [54]:

$$\mathcal{L}_{ij}(\mathbf{B}) = \beta(B) i \epsilon_{ijk} \hat{B}_k, \quad (10.7)$$

where Φ is an antisymmetric Hermitian tensor in terms of the Levi-Civita tensor ϵ_{ijk} . The

dimensionless quantity $\beta(B)$ is defined by $\beta \equiv \frac{4\pi c_0^3}{\alpha_0 \omega_0^3} \frac{V B c_0}{\gamma}$ [54].

Equation (10.2) shows that the matrix elements of the return Green function $\mathbf{G}_0(\omega)$ exhibit singularities at $r = 0$. As was explained in Chapter 2, this feature can be handled by regularization:

$$\tilde{\mathbf{G}}_0(k, \mathbf{r} = 0) = \left(\frac{\Lambda}{6\pi} + i \frac{k}{6\pi} \right) \mathbf{U}. \quad (10.8)$$

The inverse length scale Λ defines ω_0 and γ by the relations $1/\Lambda = (\omega_0/c)^2(\alpha_0/6\pi)$ and $1/\Lambda = (\gamma c/\omega_0^2)$, respectively. Using Equations (10.8) and (10.4), we can write

$$\mathbf{t}^{-1}(\omega) = \mathbf{V}^{-1}(\omega) - \mathbf{G}_0(\omega) = \frac{\Lambda}{6\pi} \left(1 - \frac{\omega_0^2}{\omega^2} \right) \mathbf{U} + i \frac{\omega}{6\pi c} - \frac{c^2}{\omega_0^2} \mathcal{L}(\mathbf{B}), \quad (10.9)$$

where Eqs. (10.5, 10.6) have been used. Assuming that $\omega_0 \gamma \ll 1$ (for an atom, typically $\omega_0 \gamma \sim 10^{-6}$) we can put, close to the resonance, $\omega \approx \omega_0$, and introduce the detuning parameter, $\Delta \equiv (\omega - \omega_0)/\gamma$. We can rewrite the total scattering matrix $\mathbf{M}(\omega)$ in (Eq. (10.3)) as:

$$\mathbf{M}(\omega) = \frac{4\pi c_0}{\omega_0} \left[\mathbf{U} \Delta + \frac{2}{3} \mathbf{U} i - \frac{4\pi c_0}{\omega_0} \mathbf{G}(\omega_0) - \mathcal{L}(\mathbf{B}) \right]^{-1}. \quad (10.10)$$

Notice that the only frequency-dependence of the total scattering matrix (10.10) near the resonance is contained in Δ . Equation (10.10) shows that to find the T -matrix (10.1), we need to diagonalize the matrix

$$\mathbf{G}_{MO}(\omega_0) = \frac{4\pi c}{\omega_0} \mathbf{G}(\omega_0) + \mathcal{L}(\mathbf{B}) \quad (10.11)$$

to find $\mathbf{M}(\omega)$ for all detunings since, quite conveniently, this matrix is *independent* of the detuning Δ . The presence of the second term in (10.11) constitutes the only difference with the standard point-dipole model [81, 82, 156, 157, 158].

In view of Eq. (10.10), we should numerically diagonalize the matrix $\mathbf{G}_{MO}$ [Eq. (10.11)] for N scatterers in order to look for the $j = 1 \dots 3N$ "proximity" resonances of the disordered

system, determined by the equation:

$$\left[\Delta + \frac{2}{3}i \right] \mathbf{U} - \zeta_j(\mathbf{B}) = 0, \quad (10.12)$$

with $\zeta_j(\mathbf{B})$ the j -th complex eigenvalue of the matrix $\mathbf{G}_{MO}$. It is important to recall that, since the system is finite and open, waves can always leak through the boundaries. The existence of resonances with finite leakage implies that the imaginary part of Eq. (10.12) must be nonvanishing and positive. As a result, the imaginary part of the eigenvalues of $\mathbf{G}_{MO}$ must obey the inequality $\text{Im}\zeta_j < 2/3$. A very small imaginary part of Eq. (10.12) (which occurs for $\text{Im}\zeta_j \rightarrow 2/3$) means that the resonance hardly leak, which would represent an "almost" localized mode. On the other hand, a situation where the imaginary part of Eq. (10.12) is negative (i.e., if $\text{Im}\zeta_j > 2/3$) can only occur if the associated resonance have gain. In order to be consistent with previous works on the point-dipole model [81, 82, 156, 157, 158], where the Green matrix is defined in a slight different way, we will define the quantity $\lambda_j(\mathbf{B}) \equiv (3i/2) \times \zeta_j(\mathbf{B})$. Rephrasing the above discussion in terms of λ_j , we notice that the real part of λ_j must obey the inequality $\text{Re}\lambda_j > -1$ if there is no gain and that $\text{Re}\lambda_j \rightarrow -1$ corresponds to an "almost" localized mode. In the following we will only refer to λ_j and analyze its distribution in the complex plane.

As an example of a typical distribution of λ 's in the complex plane for a randomly chosen $\mathbf{G}_{MO}$ matrix with vanishing magnetic field, in Fig. 10-1 we plot the real and the imaginary parts of λ corresponding to a random configuration of $N = 1000$ resonant dipoles in a sphere, with the uniform density $\rho = 1$ scatterer per wavelength cubed. This situation is equivalent to the case of scatterers without magneto-optical activity [81, 82, 156, 157, 158]. We observe that values of λ tend to "cluster" themselves around the line $\text{Re}\lambda = -1$. Previous numerical work has shown this kind of behavior by increasing the size of the system [81, 82, 156, 157, 158], suggesting that in the limit of an infinite system all λ 's will move toward the value $\text{Re}\lambda = -1$. This transition has been interpreted as the formation of a band of localized states [81, 82, 156, 157, 158], a conclusion that is consistent with the previous discussion.

The impact of magneto-optical activity on the distribution of λ 's in the complex plane can be seen in Fig. 10-2 where we plot the real and the imaginary parts of λ calculated with the

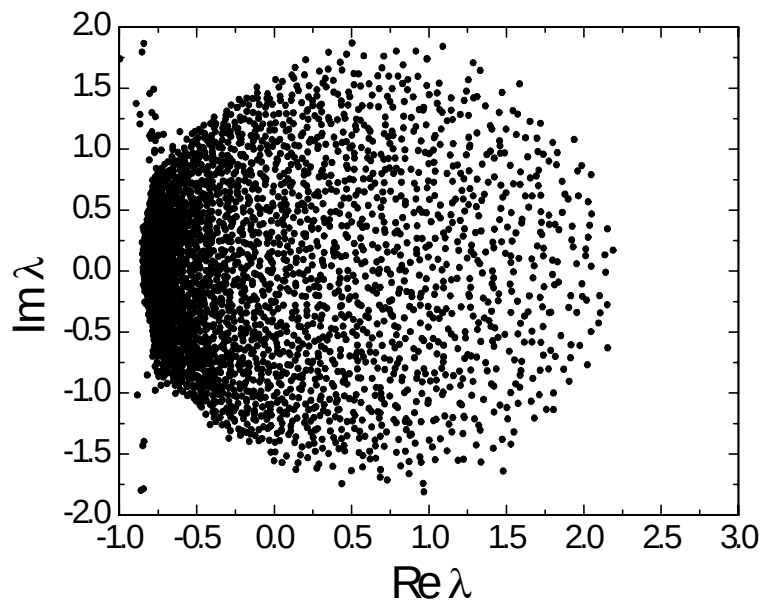


Figure 10-1: Distribution of λ 's corresponding to the spectrum of the $\mathbf{G}_{MO}$ matrix for a randomly selected configuration of $N = 1000$ pointlike scatterers calculated with a vanishing external magnetic field.

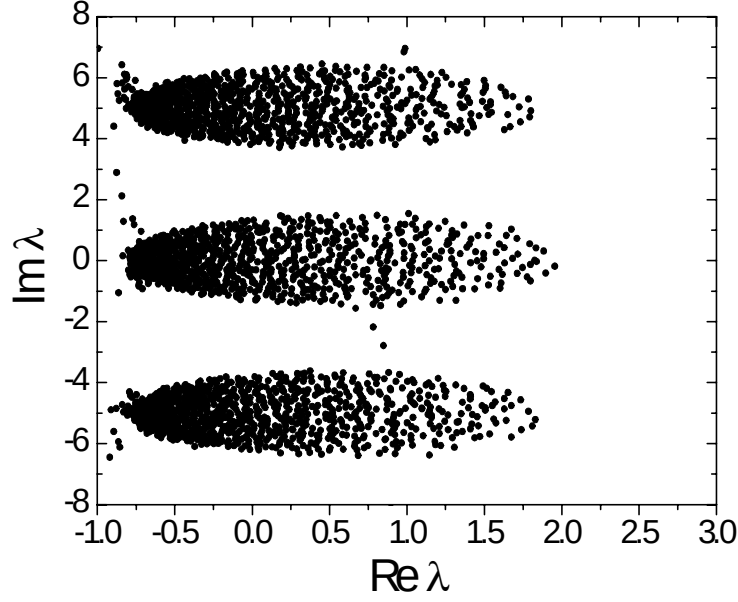


Figure 10-2: As in Fig. 10-1, but now in the presence of an external magnetic field. The value of the dimensionless magnetic field strength parameter is $\beta(B) = 5$. Notice the splitting of the values of the imaginary parts of λ , which was absent for scatterers without magneto-optical activity (see Fig. 10-1).

same parameters of Fig. 10-1 but now with a nonzero value of the external magnetic field. The value $\beta(B) = 5$ was chosen in order to allow the clear identification of the magnetic field effects on the distribution of λ 's. We not only can observe the tendency of the values of λ to "cluster" themselves around the line $\text{Re}\lambda = -1$, as was seen for scatterers without magneto-optical activity, but also the splitting its imaginary parts, $\text{Im}\lambda$. In order to explain this result, one should recall the physical meaning of the real and imaginary parts of λ . Within our model for each single scatterer, which exhibits an internal scattering resonance (also called Breit-Wigner type resonance), the real and imaginary parts of λ correspond approximately to the relative width and to the positions of the resonances, respectively [158]. As a result, the observed splitting of the values of $\text{Im}\lambda$ can be interpreted as the Zeeman splitting of the energy levels of each scatterer induced by the application of an external magnetic field.

This interpretation is confirmed by the results shown in Fig. 10-3, where we have calculated

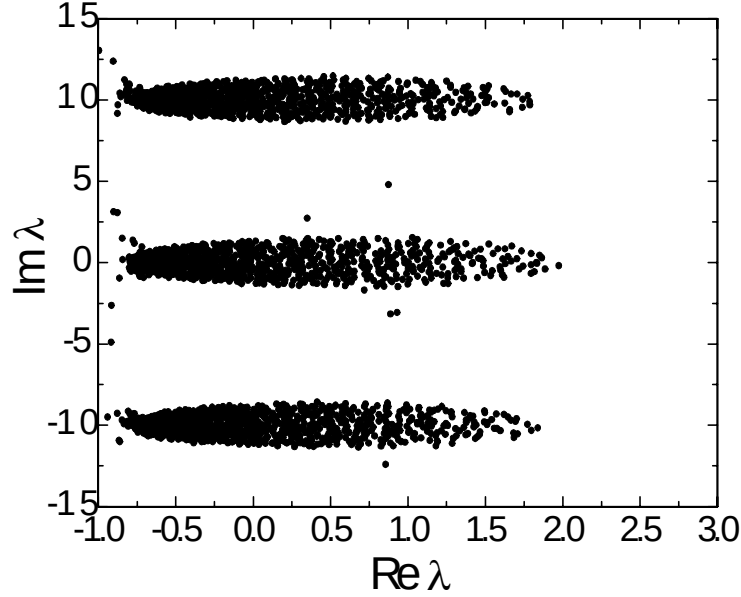


Figure 10-3: As in Fig. 10-2, but now with $\beta(B) = 10$.

the distribution of λ 's in the complex plane for the same number of scatterers and parameters used in the previous figures but now with a magnetic field strength parameter $\beta(B)$ two times larger than in Fig. 10-2, i.e., $\beta(B) = 10$. By comparing the positions where the values of $\text{Im}\lambda$ tend to distribute themselves with the ones in Fig. 10-2, we notice that they are effectively *linearly proportional* to $\beta(B)$. This fact confirms that the observed splitting of the values of $\text{Im}\lambda$ actually reflects a genuine Zeeman splitting of the energy levels of a single scatterer. In addition, we observe that for a larger value of the magnetic field, the distribution of the values of $\text{Im}\lambda$ around their splitted positions tends to be less pronounced.

In the following section, we will calculate the distribution of resonance widths $P(\Gamma)$ in the diffusive regime for magneto-optical scatterers and investigate the impact of an external magnetic field. A possible application to random lasers will also be discussed.

10.2 Distribution of resonance widths in a magnetic field: towards to a tunable random laser?

The resonance widths are calculated as in Chapter 9, i.e., using the approximation $\Gamma/\Gamma_0 \simeq 1 + \text{Re}\lambda$. Figure 10-4 exhibits the distribution of resonance widths $P(\Gamma)$ for 400 on-resonance point-dipoles randomly distributed in a sphere with uniform optical density of $\rho = 1$ scatterer per wavelength cubed (diffusive regime) with and without magnetic field (the corresponding values of the quantity $\beta(B)$ are $\beta(B) = 5$ and $\beta(B) = 0$, respectively). One can notice a region where the power law decay dominates in $P(\Gamma)$, though small, as expected for the diffusive regime. Notice also that the associated exponents do not change appreciably by the presence of the magnetic field. We have numerical evidence that a magnetic field does not change significantly the power law decay in $P(\Gamma)$ for the localized regime either, where the exponent tends towards -1 , as in the scalar case discussed in Chapter 9. For very large Γ , $P(\Gamma)$ decays faster than a power law, again as in the scalar case.

However, in the small Γ region ($\Gamma \lesssim \Gamma_T$) magnetic field effects clearly affect $P(\Gamma)$ and the peak of the distribution is slightly shifted towards larger Γ values. This change in $P(\Gamma)$ for long-living states (small Γ) suggests that prelocalized states in the diffusive regime are affected by the presence of the magnetic field. It is worth mentioning that such a change in $P(\Gamma)$ was also recently reported in numerical simulations within the kicked rotor model in the diffusive regime, in which case it has been associated with the sensibility of prelocalized states to the breaking of time-reversal symmetry [142]. As was discussed in Chapter 8, $P(\Gamma)$ for $\Gamma \lesssim \Gamma_T$ is expected to follow a log-normal distribution due the presence of prelocalized states with a factor that indeed takes into account time-reversal symmetry [see Eq. (8.7)]. Furthermore, as was argued theoretically by Mirlin using the nonlinear σ model, prelocalized states should be sensitive to time-reversal symmetry [130]. This seems to agree with our findings but a more detailed study will be necessary.

The fact that a magnetic field affects $P(\Gamma)$ for $\Gamma \lesssim \Gamma_T$ in the diffusive regime and, consequently, prelocalized states, could have important implications for the research on random lasers. As discussed in chapter 8, prelocalized states are believed to be the responsible for the observed lasing instability in samples far from the localization threshold [141] since they have

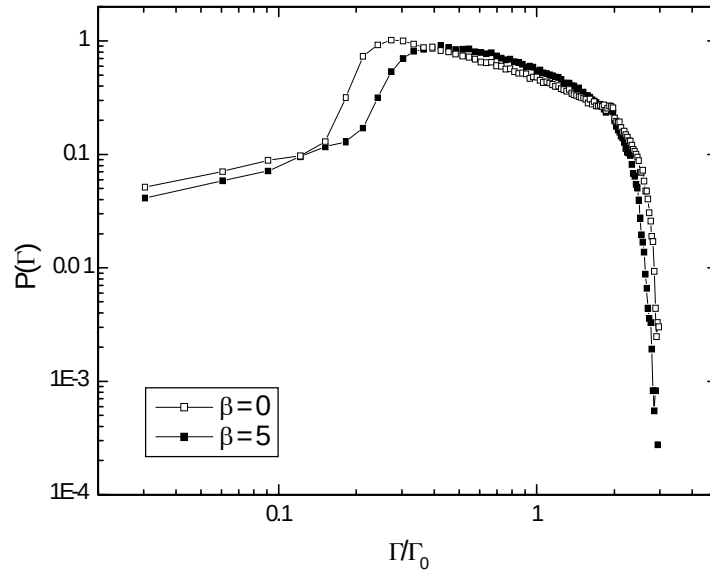


Figure 10-4: distribution of resonance widths $P(\Gamma)$ for 400 resonant point-dipoles randomly distributed in a sphere with uniform optical density of $\rho = 1$ scatterer per wavelength cubed, with and without magnetic field. The values for the dimensionless magnetic field-strength factor are: $\beta(B) = 5$ (full squares) and $\beta(B) = 0$ (open squares). The lines are just a guide for the eye.

the smallest decay rates. Therefore, one can envisage to explore the modifications induced by a magnetic field in $P(\Gamma)$ for small Γ to construct a *tunable magneto-optical random laser*. The application of an external magnetic field in a disordered medium composed of magneto-optical scatterers would modify the laser threshold of a random laser since it would affect prelocalized states as suggested by the inspection of the small Γ region in the distribution $P(\Gamma)$ shown in Fig. 10-4.

10.3 Conclusions and perspectives

In this chapter we have studied the distribution of resonance widths $P(\Gamma)$ in three-dimensions using a vector treatment for the point-dipole to describe light propagation in disordered media. Such a treatment not only constitutes a more realistic model to describe light propagation but also allows to study magneto-optical effects in random media. We have shown how to modify the point-dipole model to include magneto-optical activity inside the scatterers. It was shown that the spectra of the scattering Green matrices $\mathbf{G}_{MO}$, which characterize wave propagation in random media, is modified by the presence of an external magnetic field. This modification manifests itself by the splitting of the real parts of the eigenvalues of the Green matrix. We have numerically calculated $P(\Gamma)$ in the diffusive regime for magneto-optical dipoles. It was shown that an external magnetic field does not substantially alter the behavior of $P(\Gamma)$ neither in the intermediate Γ region (where $P(\Gamma)$ follows a power law) nor in the large Γ region, where $P(\Gamma)$ decays faster than algebraically. For the small Γ region however, the presence of a magnetic field clearly modifies the behavior of $P(\Gamma)$. We have argued that this modification can be attributed to the sensibility of prelocalized states to the breaking of time-reversal symmetry, as also put forward theoretically by Mirlin [130]. Finally, we have explored this fact to suggest that an external magnetic field could be used to design a tunable magneto-optical random laser.

Finally, let us know briefly outline some possible directions for future work. In order to get a deeper understanding of the behavior of $P(\Gamma)$ for $\Gamma \lesssim \Gamma_T$ in the diffusive regime and the importance of the prelocalized states in this region, a statistical analysis of resonance intensities would be necessary. Prelocalized states have very sharp amplitude peaks that, although being very rare among the typical states in the diffusive regime, manifest themselves in a particular

way in the far tails of the distribution of eigenstate intensities [130]. These distributions have been calculated theoretically [130], predicting even the sensibility of prelocalized states to time-reversal symmetry, and it would be interesting to compare them to numerical results obtained using the dipole-model for wave propagation. At the time of writing, the existing numerical works focusing on the far tail of distribution of the eigenstates intensities are restricted to the Anderson model, both in low-dimensional [159] and in three-dimensional media [160].

Concerning the tunable random laser, it would be desirable to change the geometry of the scattering medium in order to make the simulations more adapted to describe an actual experiment. A slab geometry would be more suited for this purpose than the spherical geometry, with which we have performed the simulations. This modification will be necessary before we can make reliable order-of-magnitude predictions for the laser threshold in a magnetic field.

Résumé du chapitre 10 : Statistique des largeurs de résonance pour les ondes vectorielles - application pour l' étude de la magnéto-optique dans les milieux aléatoires désordonnés

Ce chapitre est consacré à une extension du travail présenté au chapitre précédent pour étudier la distribution des largeurs de résonance $P(\Gamma)$ en utilisant une approche vectorielle pour la propagation des ondes électromagnétiques. Ce genre d'approche est importante pour une description plus réaliste de la propagation de la lumière dans les milieux désordonnés car elle prend en compte la polarisation des ondes. En plus, l'approche vectorielle nous permet d'étudier l'influence de la brisure de symétrie par renversement du temps, induite par la présence d'un champ magnétique externe, sur $P(\Gamma)$ et ses implications pour la localisation forte de la lumière. Nous nous intéresserons également aux possibles applications dans les lasers aléatoires.

Nous avons mis en oeuvre des modifications dans le modèle des diffuseurs ponctuelles [81][82] pour prendre en compte l'effet Faraday à l'intérieur des diffuseurs. Nous avons montré que le spectre de la matrice de Green, qui décrit la propagation des ondes dans les milieux aléatoires, est modifié par la présence du champ magnétique externe. Cette modification se manifeste par le « splitting » des parties réelles des valeurs propres de la matrice de Green.

En réalisant des simulations numériques dans ces systèmes désordonnés en régime diffus,

nous avons constaté que la présence du champ magnétique externe n'altère pas d'une façon significative la distribution $P(\Gamma)$ ni dans la région des valeurs intermédiaires de Γ ($\Gamma \gtrsim \Gamma_T$, où est la fréquence de Thouless), où $P(\Gamma)$ suit une loi de puissance, ni pour les valeurs de Γ très larges ($\Gamma \gg \Gamma_T$), où $P(\Gamma)$ décroît plus rapidement qu'une loi de puissance. Par contre, dans la région des Γ petits ($\Gamma \lesssim \Gamma_T$), la présence du champ magnétique altère substantiellement le comportement de $P(\Gamma)$. Nous attribuons cette altération à la sensibilité des états pre-localisés à la brisure de symétrie par renversement du temps, comme a été prévu théoriquement par Mirlin [130]. Finalement, nous explorons ce résultat pour suggérer que la présence d'un champ magnétique externe peut être utilisé dans le développement d'un laser aléatoire contrôlé magnéto-optiquement car le seuil de fonctionnement de ce genre de laser est typiquement donné par le mode avec le Γ le plus petit (i.e. le mode qui vit le plus longtemps) [94].

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