

# **Quantum optics and multiple scattering in dielectrics**

Cover: Picture by Yvonne Grootenboer

Printed by PrintPartners Ipskamp B.V., Enschede.

ISBN: 90-9016950-4

# **Quantum optics and multiple scattering in dielectrics**

ACADEMISCH PROEFSCHRIFT

ter verkrijging van de graad van doctor  
aan de Universiteit van Amsterdam,  
op gezag van de Rector Magnificus  
prof. mr. P.F. van der Heijden  
ten overstaan van een door het college voor promoties  
ingestelde commissie, in het openbaar te verdedigen  
in de Aula der Universiteit  
op woensdag 11 juni 2003, te 12:00 uur

door

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geboren te Hoogeveen

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The work described in this thesis is part of the research program of the  
“Stichting Fundamenteel Onderzoek der Materie” (FOM),  
which is financially supported by the  
“Nederlandse Organisatie voor Wetenschappelijk Onderzoek” (NWO).  
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## Liggen in de zon

Ik hoor het licht het zonlicht pizzicato  
de warmte spreekt weer tegen mijn gezicht  
ik lig weer dat gaat zo maar niet dat gaat zo  
ik lig weer monomaan weer monodwaas van licht.

Ik lig languit lig in mijn huid te zingen  
lig zacht te zingen antwoord op het licht  
lig dwaas zo dwaas niet buiten mensen dingen  
te zingen van het licht dat om en op mij ligt.

Ik lig hier duidelijk zeer zuidelijk lig zonder  
te weten hoe of wat ik lig alleen maar stil  
ik weet alleen het licht van wonder boven wonder  
ik weet alleen maar alles wat ik weten wil.

Hans Andreus  
Uit: *Muziek voor kijkdieren* (1951)

*Aan mijn ouders*



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# Chapter 1

## Introduction: quantum optics of photonic media

### 1.1 Light in free space and in dielectrics

We can see things because these things emit or scatter light that is subsequently absorbed in our eyes. Emission, absorption and scattering of light are examples of interactions of light with matter. According to classical optics, light is a name for those electromagnetic waves that happen to be visible for the human eye. Electromagnetic waves can be described as the solutions of Maxwell's equations. In free space, solutions of Maxwell's equations correspond to plane waves that move in straight lines. In many experiments, especially those involving interference, light appears to be a wave indeed. In other experiments, light can best be understood as a stream of particles (photons). This two-sided nature of light can nowadays be described consistently in one theory, namely in the quantum theory of light (or quantum electrodynamics). The subject of this thesis is the quantum theory of light in media other than free space. The rest of this section describes some properties of optical media that one should wish to include in such a theory.

When the field interacts with matter, the properties of the electromagnetic field are changed [1]. In air, for instance, light travels just a bit slower than in free space, because the interactions with the air molecules take some time. The factor 1.0003 by which the light is slower is the refractive index of air. If one looks even more precisely, one finds that air shows frequency dispersion: different colors move at different speeds. Violet light has a refractive index of 1.000298 and red light is faster with a refractive index of 1.000291 [2].

Another process that occurs in air but not in free space is light scattering. Air molecules and small dust particles scatter red light with long wavelengths less effectively out of sight than blue and violet light with shorter wavelengths. This explains why the sun appears red at sunset, when light waves travel a long way through air before reaching our eyes. Light scattering by air molecules is also the mechanism that explains why at daytime the sky is blue, because it is scattered light that we see. Light with shorter wavelengths is scattered more easily by air molecules towards our eyes.

Light can propagate forever in free space without disappearing, but in a medium it is possible that light gets absorbed. Absorption means that light becomes something else than light, heat for example. Light absorption is more important in fluid or solid media than in gases: sun glasses are better light absorbers than air. Sometimes the word absorption is used for scattering, but for this thesis it is important to distinguish the two processes. Both scattering and absorption can be the cause of extinction, which is loss of light in the original direction.

In general, a medium can either be described microscopically, in terms of the individual atoms or molecules, or macroscopically, with the use of a dielectric function (the square of the refractive index). In this thesis, the medium is described macroscopically. With “macroscopic” it is meant that the dielectric function is assumed constant on the atomic scale but it is allowed to vary strongly on the wavelength scale of light, as is the case in photonic media. The medium forms a modified optical environment for “guest atoms” that do not belong to the medium. Only these atoms are described microscopically. Guest atoms will interact with light and they will probe the modified optical properties [1].

Dispersion, absorption and scattering of light in a medium can be described theoretically by adding terms in the Maxwell equations. These extra terms model the pieces of matter from which the medium is built up. The modified Maxwell equations have different light waves as their solutions. Not only a light wave is different, but also a photon in a medium is different from a photon in free space. In this thesis it is studied in particular how the free-space quantum theory of light can be modified in order to describe photons in inhomogeneous media (including photonic media) or in media that show light extinction. Furthermore, calculations are presented that show strong modifications of the optical properties of media in which light strongly scatters off many scatterers.

## 1.2 Classical and quantum optics

In the past centuries scientist have debated fiercely whether light is a stream of particles or a wave. Newton developed a corpuscular theory of light (1704) while Huygens worked on his wave theory (1690). The phenomenon that two light beams can interfere destructively and constructively, can best be understood if light is a wave. Therefore, the light-as-a-wave picture had the best cards for a long time after Fresnel’s work on diffraction (1816) and its experimental verification [2]. Around 1900, it had become even almost impossible to think of light in terms of particles, because of the enormous success of the classical “Maxwell” wave theory of light [3]. And even today, classical electrodynamics suffices to explain the phenomena in many branches of optics [4].

However, observations of the photo-electric effect defied explanation around 1900: light can induce an electrical current in some metals, but only if the frequency of the light exceeds a critical value. In 1905, Einstein gave a new argument for the light-as-a-particle picture by his explanation of this effect [5]: the electrons interact with particles of light called photons. The energy  $E_\omega$  of a photon equals  $\hbar\omega$ , where  $\hbar$  is Plank’s constant divided by  $2\pi$ . Only a photon with energy high enough to overcome the binding potential of an electron, can kick it out of its bound state and make it detectable as an electrical current.

For this revolutionary explanation of the photo-electric effect Einstein received the Nobel Prize of 1921 [3].

Present-day quantum optics describes how light can have the interference properties of waves as described by Maxwell's equations, even so for light beams containing only one photon. The first experimental proof of the particle-nature of light emitted by individual atoms came relatively recently, in 1986 [6]: light was measured that was spontaneously emitted by a gas of atoms. It was found that light emitted by a single atom was detected in only one of two detectors, with equal probability. If the emitted light had been a classical wave, then one would sometimes have measured light in both detectors at the same time. The measurements proved that each single atom emitted only a single particle.

Not only the existence of photons but also the fact that they can be entangled is an important difference between classical and quantum optics. Entanglement surely is one of the mind-boggling aspects of quantum physics. An entangled system of two parts can not be treated as two separate objects with well-defined individual properties that already exist "out there" prior to measurement. A measurement on one part of the entangled system changes the state of the whole system, including the other part. Entanglement is subtle: neither the entanglement nor the measurement is some kind of interaction between the two parts. This last statement is supported by the fact that the entanglement is not influenced by the distance between the parts. Furthermore, a measurement of only one part of an entangled system never tells you that it indeed was entangled with something else; only correlation measurements of both parts tell you so.

There is a by now standard technique to entangle photons. It is based on the process of parametric down-conversion in nonlinear crystals, which gives pairs of photons with entangled momenta [7]. The electromagnetic field can also be entangled with an atom. Impressive examples can be found in cavity quantum electrodynamics, where the final state of an atom that flies through a micro-cavity depends noticeably on the presence or absence of a single photon in the cavity [8].

Many fundamental tests of quantum mechanics have been performed in the optical domain. For example, the famous Einstein-Podolsky-Rosen *Gedanken* experiments have been realized with pairs of photons with entangled polarisation directions. The measurements proved that quantum mechanical entanglement indeed exists [9].

Quantum mechanics is nowadays reconsidered and further developed with the new perspective called "quantum information theory" [7], which encompasses research in quantum computation, quantum communication and quantum cryptography. In all these areas, the idea is to put quantum entanglement to use, either for a fast and new way of computing or for inherently safe information transfer. This also drives the field of quantum optics, because photons would be suitable information carriers. Even without these (future) applications in mind, it is interesting to study the quantum theory of light in media other than free space.

## 1.3 Elements of quantum optics in dielectrics

An inhomogeneous dielectric where dispersion and absorption can be neglected, can be described by a position-dependent dielectric function  $\varepsilon(\mathbf{r})$ . The harmonic solutions of

the Maxwell equations are called optical mode functions. The mode functions  $\mathbf{f}_\lambda$  of the dielectric are solutions of the wave equation

$$-\nabla \times \nabla \times \mathbf{f}_\lambda(\mathbf{r}) + \varepsilon(\mathbf{r})(\omega_\lambda/c)^2 \mathbf{f}_\lambda(\mathbf{r}) = 0. \quad (1.1)$$

Every mode is assumed to have a different label  $\lambda$ . Light in a certain mode will forever stay in that mode, unless light interacts with matter not accounted for in the dielectric function. The latter more interesting situation presents itself when guest atoms are introduced in the dielectric that can scatter light out of one mode into another. The free-space mode functions are transverse plane waves (proportional to  $\exp(i\mathbf{k} \cdot \mathbf{r})$ ), which are the solutions of equation (1.1) in the special case that  $\varepsilon$  equals unity everywhere. These plane waves can be characterized by a wavelength, a direction, and a transverse polarization direction.

In classical optics, the amplitude and phase of the electric field are determined by a set of complex numbers, one for each mode. These numbers are the mode amplitudes. If there is no light, all mode amplitudes are zero; a larger absolute value of a particular mode amplitude means that there is more light in that mode.

In ordinary quantum mechanics, the position and momentum of a particle become operators that do not commute. These and other operators work on the quantum mechanical state vectors of the particles. In quantum optics, the electric and magnetic fields also become non-commuting operators. Again, there is a state vector and it describes the quantum state of light. The electric-field operator has the form [10–14]

$$\mathbf{E}(\mathbf{r}, t) = i \sum_{\lambda} \sqrt{\frac{\hbar \omega_{\lambda}}{2\varepsilon_0}} \left[ a_{\lambda} e^{-i\omega_{\lambda} t} \mathbf{f}_{\lambda}(\mathbf{r}) - a_{\lambda}^{\dagger} e^{i\omega_{\lambda} t} \mathbf{f}_{\lambda}^*(\mathbf{r}) \right]. \quad (1.2)$$

This mode expansion of the electric-field operator is presented already in this introductory chapter, in order to show how quantum optics describes light both in terms of waves and of particles. The optical modes in quantum optics are the same harmonic solutions  $\mathbf{f}_\lambda$  of the Maxwell equations as in classical optics. The difference with classical optics is that mode amplitudes have now been replaced by the creation and annihilation operators  $a_{\lambda}$  and  $a_{\lambda}^{\dagger}$  in Eq. (1.2). The energy operator of a particular mode is  $\hbar\omega_{\lambda}(a_{\lambda}^{\dagger}a_{\lambda} + 1/2)$ . The spectrum corresponding to a particular mode  $\lambda$  is a harmonic-oscillator spectrum with energy separation  $\hbar\omega_{\lambda}$ : the ground (or vacuum) state  $|0\rangle$  of the electromagnetic field is the state with no photons present in any mode; the  $j^{\text{th}}$  excited state of mode  $\lambda$  corresponds to  $j$  photons in that mode. Photons in a dielectric are different from free space because they are the elementary excitations of different waves than in free space.

The terms in Eq. (1.2) featuring annihilation operators taken together are called the positive-frequency part  $\mathbf{E}^{(+)}$  of the electric field; the creation-operator part is called  $\mathbf{E}^{(-)}$ . In terms of these two quantities, the light intensity operator is given by [15]

$$I(\mathbf{r}, t) = 2\varepsilon_0 c \mathbf{E}^{(-)}(\mathbf{r}, t) \cdot \mathbf{E}^{(+)}(\mathbf{r}, t). \quad (1.3)$$

A measured light intensity (in  $[J.m^{-2}.s^{-1}]$ ) corresponds to the expectation value of this operator with respect to the quantum state of light. The detector position  $\mathbf{r}$  need not be in the far field. It can be near or even inside the inhomogeneous dielectric (at a position where  $\varepsilon$  equals unity).

## 1.4 Spontaneous emission

Spontaneous emission is responsible for most of the light around us. Excited atoms will fall back to a lower energy level by emitting one or more photons. The term “spontaneous emission” was invented for this process when only the inadequate classical electromagnetic theory was around. In classical optics, if no light is present near the atom, the electric field is zero. An absent electromagnetic field can not be the external cause of the emission, which explains the word “spontaneous”. Einstein derived spontaneous-emission rates from classical theory by using energy-balance arguments, but a full understanding of spontaneous emission requires quantum optics.

In quantum optics, atoms are surrounded by an omnipresent electromagnetic field. The field is always there, even when it is in its lowest energy state. In this vacuum state, no photons are present and no light can be detected. However, the electromagnetic field fluctuates even when in the vacuum state. Similar ground-state fluctuations occur for the position and momentum of an electron in a hydrogen atom: both the electromagnetic field operators and the atomic operators obey their respective Heisenberg uncertainly relations. An excited atom feels the fluctuations of the ground-state electromagnetic field. These fluctuations cause the atom to decay “spontaneously” to a lower state. In a semiclassical picture, exactly half of the spontaneous-decay rate of an atom can be attributed to these field fluctuations; the other half of the decay rate is caused by the electric field that the atom exerts on itself, the so-called radiation reaction [16].

The process of spontaneous emission starts with an atom in an excited state and the field in the vacuum state. After the emission, the atom is in a lower-energy state and the field can be in any one-photon state that gives the same total energy before and after the emission. Usually, spontaneous-emission rates  $\Gamma$  or inverse decay times  $\tau^{-1}$  can be calculated with Fermi’s golden rule:

$$\Gamma = \frac{1}{\tau} = \left( \frac{2\pi}{\hbar^2} \right) \sum_f |\langle f | V_{AF} | i \rangle|^2 \delta(\omega_i - \omega_f). \quad (1.4)$$

The initial state  $|i\rangle$  and the final states  $|f\rangle$  are states of both the atom and the field. The delta function in the rule (1.4) ensures energy conservation. If one assumes the electric-dipole interaction between the atom and the field, one finds the free-space decay rate  $\Gamma_0 = \mu^2 \Omega^3 / (3\pi \hbar \varepsilon_0 c^3)$ , where  $\Omega$  is the relevant atomic transition frequency and  $\mu$  is the magnitude of the atomic transition dipole. In an inhomogeneous medium, the decay-rate in terms of the optical modes becomes

$$\Gamma = \frac{\pi}{\hbar \varepsilon_0} \sum_{\lambda} |\boldsymbol{\mu} \cdot \mathbf{f}_{\lambda}(\mathbf{r})|^2 \delta(\omega_{\lambda} - \Omega). \quad (1.5)$$

This formula is presented here to show that the spontaneous-decay rate in inhomogeneous dielectrics depends not only on the atomic transition frequency, but also on its dipole orientation and position in the medium. When averaged over all possible orientations of the atomic dipole moment, the position-dependent spontaneous-emission rate (1.5) becomes proportional to the local optical density of states (LDOS) at frequency  $\Omega$ . The LDOS is a classical quantity and it depends on the properties of the medium only.

## 1.5 Photonic media and photonic band gaps

Some media are designed to modify light propagation very effectively without absorbing them. These are the so-called photonic media. In order to make them, in the first place the right building blocks are needed, which must scatter light well. Secondly, the typical distance between neighboring scatterers must be of the order of the wavelength of light. Optical modes in a photonic medium differ very much from the free-space plane waves. The waves can only be calculated by taking the multiple scattering of light into account.

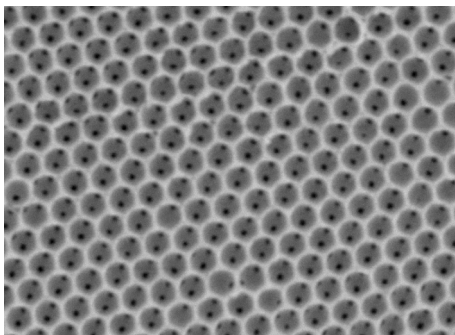
From quantum mechanics it is known that electronic band gaps exist in semiconductors or metals, because electrons Bragg-reflect off a periodic potential formed by the ions. It was proposed in 1987 to fabricate the optical analogy: a photonic band gap should exist in some three-dimensional periodic dielectric structures with a periodicity on the wavelength scale of light. These structures are also known as photonic crystals [17, 18].

It would be very interesting to have a photonic crystal with a photonic band gap: light of frequency inside the band gap will not be able to traverse the crystal in any direction and will be reflected. Even more interestingly, spontaneous emission by an atom inside the crystal would be completely suppressed. The inhibition of spontaneous emission can be illustrated with Eq. (1.5): if modes  $\lambda$  with eigenfrequencies  $\omega_\lambda$  equal to the atomic transition frequency  $\Omega$  do not exist, then the spontaneous-emission rate  $\Gamma$  is zero. Inhibition of spontaneous emission has been observed before in a medium other than a photonic crystal, namely in a cavity small compared to the optical wavelength [19]. There is no such size limitation for photonic crystals.

In model calculations of light emission in band-gap materials, many more unusual quantum optical phenomena showed up. To name a few phenomena (without explaining them here), there are predictions of anomalous Lamb shifts, localization of superradiance, oscillatory spontaneous emission rates, and fractionally populated excited states in the steady state [20–28]. For some of the phenomena, the frequency of light has to be inside the band gap, while for others it has to be just on one of the band-gap edges. Predictions of band-edge effects strongly depend on the shape and analytic behavior of the assumed optical density of states as a function of frequency [29, 30]. A photonic band-gap crystal would be a fascinating new playground for quantum electrodynamics.

The material requirements for obtaining a band gap for light are demanding. Usually, photonic crystals are two-component dielectrics with refractive indices  $n_1$  and  $n_2$ . To begin with, the material must be strongly photonic, meaning that the refractive index must vary on the wavelength scale of light and the refractive-index contrast  $n_1/n_2$  must be high. Furthermore, the volume fraction of the high-index material should be small [24, 31] and the crystals preferably should have many unit cells in all three directions. Light should not get lost by light absorption, if only because a band gap can not be defined in that case [32]. A state-of-the-art fabrication technique of three-dimensional photonic crystals is based on self-organization of colloidal spheres. Figure 1.1 shows a so-called inverted opal or air-sphere crystal [33, 34] of titania and air. Already a fivefold reduction of spontaneous-emission rates has recently been reported [35] for crystals as in figure 1.1. The search for a full photonic band gap continues until today [35–37].

Photonic crystals with refractive-index variations in only one or two dimensions do not exhibit full photonic band gaps, but they also strongly modify the propagation of

**Figure 1.1:**

Scanning-electron microscopy picture of the (111)-surface of a three-dimensional *fcc* air-sphere crystal of  $\text{TiO}_2$  (anatase) and air. The width of the picture corresponds to  $10\mu\text{m}$ . The  $\text{TiO}_2$ /air refractive index contrast is  $(2.7 \pm 0.4)$ . The air-spheres overlap each other partly. The black spots in the incomplete air spheres at the surface are the “doors” of these spheres to the complete air spheres lying one level deeper. (Picture kindly provided by L. Bechger.)

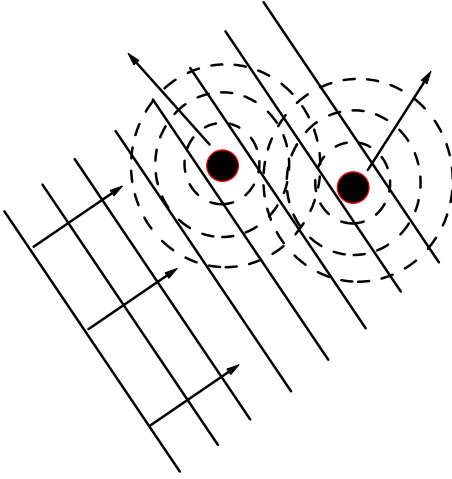
light. One-dimensional photonic crystals are also known as Bragg mirrors. Spontaneous-emission rates inside Bragg mirrors can depend strongly on the atomic position in a unit cell. Two-dimensional photonic crystals have periodic refractive-index variations in a plane. Light propagation in the in-plane directions can be fully inhibited so that light is forced to propagate in the third dimension only. This is how photonic-crystal fibres work [38].

Photonic crystals are studied both for fundamental reasons as a new playground for quantum optics and for possible applications in optical communication and opto-electronics [39]. In both cases, it is important to be able to calculate the optical properties of the realized or imagined structures.

## 1.6 Multiple light scattering

Light propagation in photonic media is in the multiple-scattering regime, and in the following it will be explained what this means. If a plane wave of light scatters off two particles, as depicted in figure 1.2, then according to Huygens’ principle each particle acts as a secondary source of light and sends out waves, which for simplicity were drawn spherical in the figure. If the particles do not scatter strongly or if they are far enough apart, then the total scattered wave is just the sum of the two waves emitted by the individual particles. This is called the single-particle scattering regime. Light scattering by air is in this regime. On the other hand, if light is scattered well by the individual scatterers and if they are close to each other, then the light scattered by the one becomes a non-negligible source of light for the other, and vice versa. Indeed, light can go back and forth many times between the two scatterers. The total scattered wave is no longer simply the sum of the two waves that the particles would have emitted if they had been alone. This is the multiple-scattering regime. Light scattering in photonic media belongs to this regime.

The (multiple) scattering properties of a medium can be inferred from the Green function  $\mathbf{G}(\mathbf{r}, \mathbf{r}', t)$  of that medium: given a light pulse  $\mathbf{E}_p(\mathbf{r}, t_0)$  at some initial time  $t_0$  before the pulse has scattered at all, the light pulse (including scattered light) at a later time at a

**Figure 1.2:**

A plane wave (denoted by parallel lines) scattered off two particles, which subsequently act as secondary sources of light. In the single-particle scattering regime, the total scattered wave is the sum of the waves scattered by individual particles alone. In the multiple-scattering regime, it is important that the particles scatter the waves scattered by the other (and so on). Then the two particles must be considered as one more complicated composite scatterer.

position  $\mathbf{r}$  is given by

$$\mathbf{E}_p(\mathbf{r}, t) = \int d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}', t - t_0) \cdot \mathbf{E}_p(\mathbf{r}', t_0). \quad (1.6)$$

This equation shows how the Green function propagates the initial field to a scattered field elsewhere in space and later in time. All information of light propagation in the medium must be included in  $\mathbf{G}$  for this relation to hold. Because of the property (1.6), one can say that the Green function is the solution of the scattering problem.

Green functions and optical mode functions  $\mathbf{f}_\lambda$  can be calculated with multiple-scattering theory. Starting with the Green function of a known medium, free space for example, as well as with the properties of the extra particles that scatter light, mode functions and the Green function of the total system can be calculated. Usually, it is easier to calculate the frequency-dependent Green function rather than the time-dependent one. The two are related by a Fourier transformation. However, some interesting properties of a multiply-scattering medium can be found from the frequency-dependent Green function, so that the often difficult inverse Fourier transformation is not needed. For example, the local density of states (LDOS), introduced in section 1.4, can be directly determined from the imaginary part of the Green function  $\mathbf{G}(\mathbf{r}, \mathbf{r}, \omega)$ .

The optical modes of an infinite photonic crystal are the solutions  $\mathbf{f}_\lambda$  of the wave equation (1.1), where the dielectric function  $\varepsilon(\mathbf{r})$  has discrete time-translation symmetry in all three directions. Because of the symmetry of the problem, the optical modes are Bloch modes which can be labelled by an incoming wave vector  $\mathbf{k}$  and a band index  $n$ . This is very much analogous to the more familiar electronic band-structure problem. The optical problem is less straightforward because the mode functions are vector functions and because the optical periodic potential is frequency-dependent. Methods for calculating band structures were recently reviewed in [40].

As was discussed in section 1.4, orientational averages of spontaneous-emission rates are proportional to the local optical density of states. The LDOS inside a photonic crystal

can not be inferred given its band structure, except for frequencies inside a band gap: in that case the LDOS simply vanishes everywhere in the unit cell. In other situations, calculations of the LDOS are numerically involved and are therefore restricted to specific positions in the unit cell [41, 42].

Real photonic crystals have finite sizes and experiments always involve crystal boundaries. The calculation of optical modes and other properties of finite crystals requires different methods. The reduced symmetry makes it natural to work in real space rather than in reciprocal space. Usually, the dielectric environment is discretized and Maxwell's equations are solved with finite-difference frequency-domain (FDFD) or time-domain (FDTD) techniques [40]. In some special cases light propagation can be analyzed further, for example for photonic crystals consisting of a finite number of parallel cylinders [43, 44].

## 1.7 True modes of a beam splitter

The equation (1.5) shows the position-dependent spontaneous-emission rate in terms of the true modes of a medium. In section 1.6 it was stated that these true modes can be calculated with multiple-scattering theory. In the following, the true modes of a relatively simple scatterer, a beam splitter, will be calculated. This example introduces the so-called T-matrix formalism of multiple-scattering theory. It is a problem in classical scattering theory to find the relation between the true modes and the free-space modes. However, scattering theory will also give relations between the operators of the true modes and the free-space operators [11].

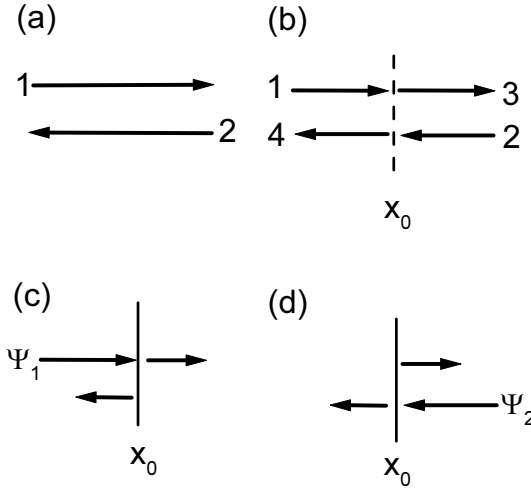
The textbook problem about a beam splitter is how to relate light in the output channels to the input-ports [15, 45]. Input-output formalisms have been set up for more complicated quantum scattering situations as well [46, 47] and these formalisms are based on formal quantum scattering theory, such as given in [48, 49]. Input-output formalisms are useful for describing non-stationary situations. The typical example is a pulse of light that enters, traverses and leaves a medium. On the other hand, the true-mode formalism is useful for calculating stationary properties such as spontaneous-emission rates inside the medium.

The present discussion of the true modes of a beam splitter starts with an even simpler situation, namely with free space. Consider light propagation in one dimension only. Assume further that the polarization direction of the light is fixed, so that light can be described as a scalar wave. Then light propagates in straight lines forever, either to the left or to the right, as sketched in figure 1.3(a). The optical modes in free space are plane waves, because plane waves are the solutions of the wave equation for light in free space:

$$[d^2/dx^2 + (\omega_k/c)^2] e^{\pm ikx} = 0. \quad (1.7)$$

The plane wave  $e^{ikx}$  corresponds to light propagating to the right, with wave vector  $k$  and frequency  $\omega_k$ ; light in mode  $e^{-ikx}$  propagates to the left. The one-dimensional analogy for the electric-field operator (1.2) is

$$E(x, t) = i \int_0^\infty dk \sqrt{\frac{\hbar\omega_k}{2(2\pi)\varepsilon_0}} \{ [a_{k,1}e^{ikx} + a_{k,2}e^{-ikx}] e^{-i\omega_k t} - \text{H.c.} \}. \quad (1.8)$$

**Figure 1.3:**

Sketches of optical modes in one dimension. In figure (a), the arrow to the right denotes the plane-wave mode  $\exp(ikx)$  and the other arrow to the mode  $\exp(-ikx)$ . In figure (b), each mode of figure (a) is split into two new modes, corresponding to light propagating to the left or to the right of the imaginary cut at position  $x_0$ . There are two input channels (1 and 2) and two output channels (3 and 4). In figure (c) and (d), a beam splitter is placed at position  $x_0$  that reflects part of the light. Figure (c) shows the “true” mode  $\psi_1$ , which comes in from the left and is partially reflected and partially transmitted. Similarly, the true mode  $\psi_2$  represented in figure (d) corresponds to light coming from the right.

For later use, the free-space Green function  $g_0$  will be introduced as the solution of the same wave equation (1.7), but with a delta-function source at the right-hand side of the equation:

$$\left[ \frac{d^2}{dx^2} + (\omega_k/c)^2 \right] g_0(x, x', \omega) = \delta(x - x'), \quad (1.9)$$

where the right-hand side is a Dirac delta function. It can be checked that  $g_0(x, x', \omega)$  is equal to  $\exp(i\omega^+|x - x'|/c)/[2i(\omega/c)]$ , where  $\omega^+$  equals  $(\omega + i\eta)$  and  $\eta$  is infinitesimally small and positive.

Suppose now that in free space one would like to measure light in a plane wave-mode, either at a position to the left of some arbitrary position  $x_0$ , or to the right of it. Suppose that the detectors are sensitive to the direction in which the photon is heading. The situation is sketched in figure 1.3(b): there are four modes corresponding to light moving to the left or right, at the left or right of  $x_0$ . The first mode function is a plane wave moving to the right, proportional to  $\exp(ikx)$ , when  $x$  is smaller than  $x_0$  and it is zero for larger  $x$ . This mode function can be summarized as  $\theta(x_0 - x)\sqrt{2}\exp(ikx)$ ; the factor  $\sqrt{2}$  gives the new mode function the same norm as the mode  $\exp(ikx)$  on the whole real axis. The other three mode functions can be written down analogously. The description of light in terms of the four half-space modes is equivalent to the two full-space plane-wave modes that

we started with. In the electric-field operator (1.8), the only thing that changes in the new representation is the quantity between square brackets, which now becomes

$$\sqrt{2} [\theta(x_0 - x)(b_{k,1}e^{ikx} + b_{k,4}e^{-ikx}) + \theta(x - x_0)(b_{k,2}e^{-ikx} + b_{k,3}e^{ikx})]. \quad (1.10)$$

Now add a beam splitter at position  $x_0$ . The presence of the beam splitter can be modelled as a delta-function potential of strength  $V$  in the wave equation for the optical modes. The wave equation changes from Eq. (1.7) into

$$\{d^2/dx^2 + [(\omega_k/c)^2 - V\delta(x - x_0)]\} \psi_{k,1}(x) = 0. \quad (1.11)$$

The harmonic solutions of this equation no longer are plane waves, but the unknown mode functions  $\psi_{k,1}$  and  $\psi_{k,2}$ , corresponding to light coming from the left or right, respectively. As an *Ansatz*, suppose that the unknown modes have the form

$$\psi_{k,1}(x) = e^{ikx} + g_0(x, x_0, \omega)T(\omega)e^{ikx_0}. \quad (1.12a)$$

$$\psi_{k',2}(x) = e^{-ik'x} + g_0(x, x_0, \omega')T(\omega')e^{-ik'x_0}, \quad (1.12b)$$

where the Green function (1.9) shows up. (The reason why the unknown modes are supposed to have this form will be given in chapter 2.) The new terms at the right-hand sides can be read from right to left: at position  $x_0$ , the plane wave scatters as described by the unknown quantity  $T(\omega)$ . Finally, the free-space Green function describes how scattered light propagates away from the beam splitter. The unknown quantity  $T(\omega)$  is called the T-matrix of the beam splitter. With the use of Eqs. (1.7), (1.11) and the definition (1.9) of the free-space Green function, one can check that the Ansatz (1.12a) indeed gives two exact solutions of the wave equation (1.11), if

$$T(\omega) = V/[1 - g_0(x_0, x_0, \omega)V]. \quad (1.13)$$

The T-matrix contains powers of the potential  $V$  up to infinite order, as one can see by expanding  $(1 - g_0V)^{-1}$  into an infinite series. The T-matrix can thus be seen as an exact summation of an infinitely long perturbation expansion in terms of the potential  $V$ . The mode function  $\psi_1(x)$  corresponds to a plane wave coming from the left that is partially reflected and partially transmitted, as in figure 1.3(c). Figure 1.3(d) depicts the mode function  $\psi_2$  that corresponds to a plane wave coming from the right.

The free-space plane-wave modes are independent, in the sense that their inner product  $\int dx [\exp(ikx)]^* \exp(ik'x)$  is zero for  $k$  unequal to  $k'$ . Physically, this means that in free space light in one mode will never jump to another plane-wave mode. From now on assume that  $x_0 \equiv 0$ . Now consider the inner product of the two true modes  $\psi_1$  and  $\psi_2$  of the beam splitter:

$$\begin{aligned} \langle \psi_{k,1} | \psi_{k',2} \rangle &= \int_{-\infty}^{\infty} dx \psi_{k,1}^*(x) \psi_{k',2}(x) \\ &= 2\pi\delta(k - k') [1 + \text{Re}(W) + |W|^2] + 2\pi\delta(k + k')\text{Re}(W), \end{aligned} \quad (1.14)$$

where  $W$  is an abbreviation for  $T/[2i(\omega/c)]$ . The wave vectors  $k$  and  $k'$  are both positive, so that the term proportional to  $\delta(k + k')$  does not contribute to the inner product. Then it

follows that the inner product (1.14) of the true modes is equal to the inner product of the free-space plane wave modes if  $[\text{Re}(W) + |W|^2]$  is equal to zero, in other words if

$$\text{Im } T(\omega) = -\frac{|T(\omega)|^2}{2(\omega/c)}. \quad (1.15)$$

This nonlinear relation of the T-matrix is called the optical theorem of the beam splitter. The theorem only holds if the potential  $V$  representing the beam splitter is a real quantity. This statement can be checked by inserting the form (1.13) of the T-matrix into the optical theorem (1.15). A potential  $V$  with an imaginary part would model loss or gain of light in the beam splitter. Thus, the true modes are orthonormal if the beam splitter scatters light elastically.

In the presence of the beam splitter, the electric-field operator (1.8) can be expanded in terms of the true modes instead. All that changes in Eq. (1.8) is the term between the square brackets, which becomes

$$[c_{k,1}\psi_{k,1}(x) + c_{k,2}\psi_{k,2}(x)], \quad (1.16)$$

where  $c_{k,1}$  and  $c_{k,2}$  are annihilation operators corresponding to the true modes. For each mode separately, the commutation relations of the creation and annihilation operators are just like in the standard quantum mechanical treatment of the harmonic oscillator:  $[c, c^\dagger] = 1$ ; operators that belong to different modes commute.

One could measure light to the left or to the right of  $x_0 = 0$ , and heading towards or away from the beam splitter. The electric-field operators (1.16) in terms of the true modes and (1.10) in terms of the half-space modes should therefore be equivalent. By taking the inner products with  $\psi_{k',1}$  and  $\psi_{k',2}$  of the assumed equality, one finds that the equivalence only holds if

$$c_{k,1} = [b_{k,1} + (1 + W^*)b_{k,3} + W^*b_{k,4}]/\sqrt{2}, \quad (1.17a)$$

$$c_{k,2} = [b_{k,2} + W^*b_{k,3} + (1 + W^*)b_{k,4}]/\sqrt{2}. \quad (1.17b)$$

One can check that these relations between operators in different mode expansions can only be consistent with the standard commutation relations of both the operators  $c$  and  $b$  in each expansion, if the optical theorem (1.15) holds.

The example of the beam splitter served to show that an optical theorem holds if light is scattered elastically; that the theorem guarantees that true modes have the same orthonormality relations as the original modes without the scatterers. Finally, the importance of the optical theorem in quantum optics is that it ensures that mode operators of true modes can consistently be written in terms of the original operators, as shown above.

If the optical theorem does not hold, in the case of absorption or gain in the beam splitter, then the quantum optical formalism has to be modified. The Kramers-Kronig relations tell that in general, the dielectric function is a complex quantity as a function of frequency [50, 51]. So one can always find frequencies for which a beam splitter absorbs a fraction of the light. Quantum optical descriptions of light propagation in absorbing dielectrics rely on the so-called fluctuation-dissipation theorem: wherever there is absorption, there are also sources of quantum noise. These noise sources show up in operator relations like (1.17a) and (1.17b) so as to keep the relations consistent when there is loss [52].

## 1.8 Outlook into this thesis

In the chapters 2 and 3 of this thesis, it is shown how a layered dielectric (or dielectric mirror) can be modelled as a crystal of infinitely thin planes. Multiple-scattering theory is used to calculate the propagating and guided modes of this finite one-dimensional photonic crystal. The formalism allows a relatively easy calculation of the Green function of such a structure. It is studied how the spontaneous-emission rate of a radiating atom depends on the atomic position and dipole orientation. Chapter 2 deals with scalar waves. The  $T$  matrix formalism of multiple-scattering theory is introduced. In chapter 3 the formalism is generalized to vector waves in order to study the influence of dipole orientation on spontaneous-emission rates. The free-space Green function must be modified (“regularized”) in order to set up the T-matrix formalism for vector waves.

The subject of chapter 4 is the quantum optical description of light in inhomogeneous dielectrics, and the interaction of guest atoms with light. The chapter is quite formal and the dielectric function  $\epsilon(\mathbf{r})$  is left arbitrary. Material dispersion and loss of the dielectric are neglected. Models of finite or infinite photonic crystals with a periodic variation of the refractive index  $n(\mathbf{r})$  belong to this class. Starting from a minimal-coupling Lagrangian, a Hamiltonian is derived with multipolar interaction between light and the guest atoms. Special attention is paid to the derivation of Maxwell’s equations after choosing a suitable gauge in which all (static and retarded) interactions between atoms are mediated by the electromagnetic field. In the dipole approximation, it is found that a dipole couples to a field that at first sight is neither the electric nor the displacement field. Results of this chapter justify the calculations of spontaneous-emission rates of chapters 2 and 3. Furthermore, the results obtained here are the starting point for chapter 5.

Single-atom decay rates change in the presence of a dielectric, but also multi-atom processes such as superradiance will be modified. This is the subject of chapter 5. The mode functions and Green function of the dielectric are assumed to be known, determined perhaps with the use of multiple-scattering theory as in chapters 2 and 3. The T-matrix formalism of multiple-scattering theory is applied here to study the single- and multi-atom processes of guest atoms in the dielectric. Spontaneous-emission rates and elastic scattering of single atoms are considered. The strength of the formalism lies in the fact that results can readily be generalized to more than one guest atom. This is shown in the canonical example of two-atom superradiance in an inhomogeneous dielectric.

Finally, in chapter 6, the effects of material dispersion and absorption on spontaneous-emission rates in a homogeneous dielectric are considered. In a damped-polariton model for the dielectric, light is coupled to a material resonance, which in turn is coupled to a continuum into which electromagnetic energy can dissipate (or scatter, if the electromagnetic field serves as the continuum). From the microscopic model, a complex dielectric function  $\epsilon(\omega)$  is obtained that satisfies the Kramers-Kronig relations. Different dielectric functions can be obtained, depending on the interactions between light and matter and between matter and continuum. Maxwell field operators are obtained in a convenient way and their form justifies more phenomenological approaches. The formalism is used to study time-dependent spontaneous-emission rates near material resonances, in a frequency range where the optical density of states changes rapidly.



## Chapter 2

# Scalar waves in finite crystals of plane scatterers

*Spontaneous-emission rates in a dielectric structure are position-dependent and proportional to the local optical density of states. The latter can be calculated when the Green function is known. Here, dielectric slabs are modelled as infinitely thin planes and light is simplified as a scalar wave. The T-matrix formalism of multiple-scattering theory is used to find an exact analytical expression for the Green function of a dielectric mirror of arbitrary number of unit cells  $N$ . All the propagating and guided modes of the structure are found. There are at most  $N$  guided modes and their dispersion relations are studied. The guided modes appear around frequencies corresponding to the first stop band in normal direction. Local densities of propagating and guided modes are presented, also for frequencies where the layered structure acts as an omni-directional mirror.*

### 2.1 Introduction

In inhomogeneous materials like photonic crystals, spontaneous-emission rates in general are position-dependent and proportional to the local density of states (LDOS) [53, 54]. It is therefore interesting to calculate the LDOS for specific dielectric geometries. Some recent work in this area was already mentioned in the introductory chapter 1.

In principle, the LDOS can directly be inferred from the Green function of a dielectric. However, in many cases Green functions are hard or practically impossible to find, for example for finite photonic crystals with refractive-index variations in all three dimensions. Therefore, it is interesting to look for finite photonic systems for which Green functions can be calculated explicitly. In this chapter, three-dimensional light propagation is studied for a finite system of periodic dielectric layers, where the refractive index varies in one spatial direction only. The focus will be on the local densities of states in and around the structure. We look for a model that is simple enough to allow for analytical solutions yet realistic enough as to give insight.

Before introducing our model, we briefly discuss some of the relevant work on layered photonic crystals. Optical properties of dielectric slab structures have been studied intensively [2, 55], both theoretically and experimentally in pure and applied optics [56, 57]. An optically excited atom inside a slab structure can spontaneously emit its photon into a radiative or a guided mode and both types of decay channels contribute to the total spontaneous-emission rate [58]. Layered dielectric structures that recently enjoyed some interest are the so-called “omni-directional mirrors” [59]. With the future photonic band-gap crystals they share the property of omni-directional reflection of light in a well-defined frequency interval. Unlike inside a photonic band gap crystal, inside layered omni-directional mirrors the spontaneous-emission rates do not vanish [60]: in the frequency interval where light can not enter the structure, spontaneous emission is taken over by the guided modes. Evidently, reflection experiments alone are not enough to tell what is going on inside a photonic crystal.

The perfect periodicity of infinite crystals increases the chances for analytical solutions. Analytical properties of finite crystals consisting of  $N$  unit cells are more difficult to find. Fortunately it is known from transfer matrix techniques [2] how to express transmission and reflection of  $N$ -period Bragg mirrors simply in terms of the number  $N$  and the reflection  $\rho$  of a single unit cell [55]. In [61] this result was generalized to non-absorbing layered structures where the unit cell has an arbitrary refractive index profile. These analytical results are important. But as we learnt from the omni-directional mirror, the knowledge of transmission and reflection is not enough for understanding spontaneous-emission rates of internal sources: we also need to find the guided modes of finite one-dimensional photonic crystals consisting of an arbitrary number of unit cells. In [62] a general method was derived to compute propagating (or radiative) and guided modes and Green functions of multilayer dielectrics with constant refractive indices in each layer. One has to be very careful not to miss any of the guided modes. The method involves a numerical step, namely the solution of boundary conditions at each dielectric interface. In our calculations, we want to avoid such a step.

In the theory of (metallic) diffraction gratings, it proved useful to study the limit of infinitely thin gratings [63] and there homogeneous layers are called impedance sheets. We study a Kronig-Penney model of a finite layered photonic crystal. Such models have been used before in the study of infinite layered photonic crystals in one [64] and two [31, 65] spatial dimensions; in the latter article also guided modes of the infinite crystal are considered. Our model differs from these works in that we consider finite crystals in three spatial dimensions, in a formalism that is particularly suited for finding all the optical modes in a unified way; the (local) density of states strongly depends on the number of spatial dimensions considered [42].

We use the T-matrix formalism of multiple-scattering theory (see the classics [48, 66] or [67] for a recent account.) An advantage of multiple-scattering theory is that the properties of systems consisting of many scatterers can be given in terms of the properties of the individual scatterers. Boundary conditions are automatically satisfied if they are satisfied by the single scatterer. Another advantage is that the formalism simplifies the calculation of Green functions enormously. The concept of a point scatterer has proved extremely useful for random multiple scattering [68, 69]. Band structures and local densities of states

of crystals consisting of point scatterers have also been studied [53, 70]. Here we propose the planar analogue of the point scatterer, the infinitely thin plane, which we coin the *plane scatterer*. In this chapter, we consider the scattering of planes by scalar waves and in chapter 3 vector wave scattering is studied.

The organization of the chapter is as follows: in section 2.2 we introduce the multiple-scattering formalism which we then apply to a plane scatterers in section 2.3, in particular to a single plane scatterer in section 2.3.1 and to  $N$  plane scatterers at arbitrary positions in section 2.3.2. An analytical expression for the T-matrix is derived in the special case that the  $N$  planes are at regular distances. The specific model for a plane scatterer that will be used throughout this chapter is introduced in section 2.3.3. The optical modes of the crystal of plane scatterers are presented in section 2.4 and both the propagating (section 2.4.1) and the guided modes (section 2.4.2) are discussed. In section 2.5 the local densities of states corresponding to both propagating modes and guided modes are presented. In section 2.6 we conclude.

## 2.2 Multiple-scattering theory for scalar waves

The wave equation for the scalar field  $\psi_0(\mathbf{r}, \omega)$  in vacuum is

$$[\nabla^2 + (\omega/c)^2] \psi_0(\mathbf{r}, \omega) = 0. \quad (2.1)$$

Here and in the following, the frequency  $\omega$  is understood to contain an infinitesimally small positive imaginary part. Hereby we choose the causal solutions of the wave equation. The solutions of Eq. (2.1) are plane waves with wave vector  $\mathbf{k}$ . With the wave equation (2.1) the free-space Green function is associated that satisfies

$$[\nabla^2 + (\omega/c)^2] g_0(\mathbf{r}, \mathbf{r}', \omega) = \delta^3(\mathbf{r} - \mathbf{r}'). \quad (2.2)$$

Now it will be convenient to consider Eq. (2.1) as the real-space representation of an abstract tensor operator  $L(\omega)$  working on the vector field  $|\psi_0(\omega)\rangle$ , so that (2.1) in abstract notation becomes

$$L(\omega)|\psi_0(\omega)\rangle = 0. \quad (2.3)$$

The advantages of abstracting from a specific representation are that formulas look simpler, that it makes you realize that the identities do not depend on a specific representation, and that in a later stage a representation can be chosen in accordance with the spatial symmetry of the dielectric under study. The abstract vector field  $|\psi_0(\omega)\rangle$  is analogous to a quantum mechanical *state*  $|\Psi\rangle$ , whereas the real-space representation of the field  $\psi_0(\mathbf{r}, \omega) \equiv \langle \mathbf{r} | \psi_0(\omega) \rangle$  corresponds to the *wave function*  $\Psi(\mathbf{r})$ . In the same notation the Green function  $g_0(\omega)$  satisfies

$$L(\omega) \cdot g_0(\omega) = \mathbb{1} \quad (2.4)$$

Here,  $\mathbb{1}$  is the identity operator in real space and it has the property  $\langle \mathbf{r} | \mathbb{1} | \mathbf{r}' \rangle = \delta^3(\mathbf{r} - \mathbf{r}')$ .

For an inhomogeneous dispersive linear dielectric, the wave equation for the scalar field is modified as follows:

$$[\nabla^2 + (\omega/c)^2] \psi(\mathbf{r}, \omega) = -[(\varepsilon(\mathbf{r}, \omega) - 1)(\omega/c)^2] \psi(\mathbf{r}, \omega) \equiv V(\mathbf{r}, \omega)\psi(\mathbf{r}, \omega), \quad (2.5)$$

where in the last equality the frequency-dependent optical potential  $V$  was defined in terms of the dielectric function  $\varepsilon(\mathbf{r}, \omega)$ . We want to solve the “scattering problem”, meaning that we want to know how the scalar field in vacuum changes due to the presence of the dielectric. The scalar field  $|\psi_{\text{in}}(\omega)\rangle$  is modified into  $|\psi(\omega)\rangle$ , and the two fields are related through the Lippmann-Schwinger equation (see reference [71])

$$|\psi(\omega)\rangle = |\psi_{\text{in}}(\omega)\rangle + g_0(\omega)V(\omega)|\psi(\omega)\rangle. \quad (2.6)$$

The field  $|\psi(\omega)\rangle$  that satisfies Eq. (2.6) is also a solution of Eq. (2.5). The advantage of the Lippmann-Schwinger equation is that the solution can be found iteratively in higher and higher orders of the optical potential  $V$ :

$$|\psi\rangle = |\psi_{\text{in}}\rangle + g_0V|\psi_{\text{in}}\rangle + g_0Vg_0V|\psi_{\text{in}}\rangle + \dots \quad (2.7)$$

So we have a multiple-scattering series for the scalar field in terms of the potential, both when the potential consists of many scatterers and when it models just a single scatterer. (In other words, multiple-scattering theory does not require more than one scatterer.) A T-matrix can be defined that sums up how the field depends on incoming field and scatterer:

$$|\psi(\omega)\rangle = |\psi_{\text{in}}(\omega)\rangle + g_0(\omega)T(\omega)|\psi_{\text{in}}(\omega)\rangle. \quad (2.8)$$

This is a formal definition of the T-matrix, and by combining Eqs. (2.7) and (2.8), the formal solution for the T-matrix is

$$T(\omega) = V(\omega) [1 - g_0(\omega)V(\omega)]^{-1}. \quad (2.9)$$

Note that the T-matrix does not depend on the incoming field, which reflects our choice of only considering polarizabilities of the dielectric that are linear in the incoming field. It is clear that the scattering problem is solved exactly once the T-matrix is known. If an explicit form of the T-matrix can not be found, then at least we can make a first order approximation in the interaction potential  $V(\omega)$  for the T-matrix. This gives  $T(\omega) \simeq V(\omega)$ . We find the field in the well-known Born approximation when inserting this approximate result into equation (2.8).

The presence of the scatterer not only modifies already existing field modes from  $\psi_{\text{in}}$  into  $\psi$ , but it can also call into existence new modes that are bound to the scatterer. In electromagnetism, this means that light is confined in at least one of the three spatial directions. Such bound modes do not correspond to an incident field and consequently they are solutions of the Lippmann-Schwinger equation in the absence of an incident field:

$$|\psi(\omega)\rangle = g_0(\omega)V(\omega)|\psi(\omega)\rangle. \quad (2.10)$$

With the help of the formal definition (2.9) of the T-matrix, we can rewrite this homogeneous equation as

$$V(\omega)T^{-1}(\omega)|\psi(\omega)\rangle = 0. \quad (2.11)$$

This equation shows that nontrivial bound solutions of the scalar field will correspond to the poles of the T-matrix. So the  $T$  matrix not only solves the scattering problem for incident fields but also contains all information about bound modes.

In the presence of the dielectric the Green function also changes from  $g_0$  to  $g$  and the latter satisfies the following equation

$$[L(\omega) - V(\omega)]g(\omega) = \mathbb{1}, \quad (2.12)$$

The solution for the Green function analogous to Eq. (2.6) for the scalar field is the Dyson-Schwinger equation

$$g(\omega) = g_0(\omega) + g_0(\omega)V(\omega)g(\omega). \quad (2.13)$$

It can be verified that a solution of (2.13) also is a solution of equation (2.12). The problem how to find such a solution is solved once the T-matrix (2.9) is determined, because an iteration of Eq. (2.13) analogous to the series expansion (2.7) for the scalar field shows that the Green function can also be expressed in terms of the T-matrix:

$$g(\omega) = g_0(\omega) + g_0(\omega)T(\omega)g_0(\omega). \quad (2.14)$$

For most types of dielectrics the T-matrix can not be calculated explicitly, not for a single scatterer let alone for a collection of them, but we will concentrate on physically interesting situations where explicit solutions can be found. If there is more than one scatterer, it is useful to first determine the single-scatterer T-matrices for the individual scatterers. The next step is to find the T-matrix for the whole collection of scatterers in terms of the single-plane scatterers. The equations (2.13) and (2.14) also hold when the total potential  $V(\omega)$  is a sum of single-scatterer potentials  $V_\alpha(\omega)$ . By iterating one finds that the total T-matrix for an arbitrary number  $N$  of these scatterers is

$$T^{(N)} = \sum_{\alpha} V_{\alpha} + \sum_{\beta} \sum_{\alpha} V_{\beta} g_0 V_{\alpha} + \sum_{\gamma} \sum_{\beta} \sum_{\alpha} V_{\gamma} g_0 V_{\beta} g_0 V_{\alpha} + \dots \quad (2.15)$$

However, instead of expanding the total T-matrix in terms of powers of single-plane potentials, it is much more convenient to do the expansion in terms of the single-plane T-matrices:

$$T^{(N)} = \sum_{\alpha} T_{\alpha} + \sum_{\beta \neq \alpha} \sum_{\alpha} T_{\beta} g_0 T_{\alpha} + \sum_{\gamma \neq \beta} \sum_{\beta \neq \alpha} \sum_{\alpha} T_{\gamma} g_0 T_{\beta} g_0 T_{\alpha} + \dots \quad (2.16)$$

The equivalence of both expansions can be checked by expanding the single-plane T-matrices in the last equation in terms of the single-plane potentials. The expansion (2.16) is to be preferred to (2.15) when leaving out higher-order terms in the expansion, since in doing so in (2.16) the multiple-scattering by a single scatterer has been incorporated exactly, whereas by truncating the series in (2.15) also multiple scattering by single scatterers is approximated. Furthermore, in the expansion (2.16) it is more obvious whether the series can be summed explicitly or not.

This formal section on scattering theory can be summarized by stressing how useful T-matrices can be. The concept of a  $T$  matrix was introduced and it was shown how the T-matrix entails an exact solution of a scattering problem both for scattered and for bound modes of light. The same T-matrix also leads to the Green function of the dielectric system in the presence of the scatterer. Finally, it was explained why single-scatterer T matrices are useful for setting up a many-scatterer multiple-scattering theory.

## 2.3 T-matrix formalism for plane scatterers

### 2.3.1 General properties of the single plane scatterer

In this chapter we consider scattering of *scalar* waves. The situation is more complicated for vector waves, as we will see in chapter 3. Now consider an infinitely thin plane  $z = z_\alpha$  that scatters light coming from  $z < z_\alpha$ . If an incident plane wave  $|\psi_{\text{in}}\rangle = |\mathbf{k}\rangle = |\mathbf{k}_\parallel, k_z\rangle$  with  $k_z$  real and  $> 0$  scatters from the plane, the in-plane wave vector  $\mathbf{k}_\parallel$  is conserved. The total wave  $|\psi_{\mathbf{k}}\rangle$  is called a ‘true mode’ or ‘mode function’ and it has an incoming, a transmitted and a reflected part. The T-matrix for such a plane scatterer must have the form

$$T_\alpha(\omega) = \frac{1}{(2\pi)^2} \int d^2\mathbf{k}_\parallel T(\mathbf{k}_\parallel, \omega) |\mathbf{k}_\parallel, z_\alpha\rangle \langle z_\alpha, \mathbf{k}_\parallel|. \quad (2.17)$$

The function  $T(\mathbf{k}_\parallel, \omega)$  describes the light scattering as a function of in-plane wave vector and frequency. At the moment the function is not specified, but in section 2.3.3 a model will be derived for it. The T-matrix formalism should not be confused with the transfer matrix approach, which in the study of layered systems is most commonly used [2, 55]. In the rest of this section, we express transmission and reflection properties of a single plane in terms of its T-matrix. Furthermore, we analyze which forms the T-matrix can take if we assume that energy is conserved in the scattering process.

First we need a suitable representation for the plane-scattering problem. The free-space Green function  $g_0(\omega)$  has the well-known real space representation  $g_0(\mathbf{r}, \mathbf{r}', \omega) = -\exp(i\omega^+|\mathbf{r} - \mathbf{r}'|)/(4\pi|\mathbf{r} - \mathbf{r}'|)$  with  $\omega^+ \equiv \omega + i\eta$  so that the Green function corresponds to an outgoing spherical wave. In Fourier representation we have  $g_0(\mathbf{k}, \mathbf{k}', \omega) = (2\pi)^3 \delta^3(\mathbf{k} - \mathbf{k}')/[(\omega^+/c)^2 - k^2]$ . By the choice of the infinitesimally small positive part we select the causal Green function whose inverse Fourier transform gives zero for times  $t < 0$  [72]. In the following, Green functions are implicitly understood to be causal and the ‘+’-notation is dropped. Now the form (2.17) of the plane scatterer suggests a mixed representation  $\{|\mathbf{k}_\parallel, z\rangle\}$  where  $\mathbf{k}_\parallel \equiv (k_x, k_y)$  is the wave vector parallel to the planes. Then the free-space Green function becomes

$$g_0(\mathbf{k}_\parallel, z, \mathbf{k}'_\parallel, z', \omega) \equiv \langle \mathbf{k}_\parallel, z | g_0(\omega) | \mathbf{k}'_\parallel, z' \rangle = (2\pi)^2 \delta^2(\mathbf{k}_\parallel - \mathbf{k}'_\parallel) g_0(\mathbf{k}_\parallel, z - z', \omega). \quad (2.18)$$

The function  $g_0(\mathbf{k}_\parallel, z, \omega)$  will also be called the Green function and it takes the form

$$g_0(\mathbf{k}_\parallel, z, \omega) = \frac{\exp(ik_z|z|)}{2ik_z}, \quad (2.19)$$

where  $k_z = \sqrt{(\omega/c)^2 - k_\parallel^2}$  can be real or imaginary. Using this and equations (2.8) and (2.17), we find for the mode function

$$\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k} \cdot \mathbf{r}} \left[ 1 + \frac{T(\mathbf{k}_\parallel, \omega)}{2ik_z} \right], \quad (z > z_\alpha) \quad (2.20a)$$

$$= e^{i\mathbf{k} \cdot \mathbf{r}} + e^{i\mathbf{k}^R \cdot \mathbf{r}} \left[ \frac{T(\mathbf{k}_\parallel, \omega) e^{2ik_z z_\alpha}}{2ik_z} \right] \quad (z < z_\alpha). \quad (2.20b)$$

The expression between the square brackets in (2.20a) is the transmission amplitude  $\tau(\mathbf{k}_{\parallel}, \omega)$  in terms of the T-matrix and between the brackets in (2.20b) we have the reflection amplitude  $\rho(\mathbf{k}_{\parallel}, \omega)$ . The reflected wave vector  $\mathbf{k}^R$  equals  $(\mathbf{k}_{\parallel}, -k_z)$ . Thus, we have expressed transmission and reflection properties of a single plane in terms of its T-matrix.

Until now, we have not assumed that energy is conserved in the scattering process. Energy conservation puts a restriction on the form that  $T(\mathbf{k}_{\parallel}, \omega)$  can have, a restriction which we will now derive. Energy conservation can be quantified with the help of the energy flux density vector  $\mathbf{J}$  [45]:

$$\mathbf{J}(\mathbf{r}, t) = -\alpha \left[ \frac{\partial \psi^*(\mathbf{r}, t)}{\partial t} \nabla \psi(\mathbf{r}, t) + \frac{\partial \psi(\mathbf{r}, t)}{\partial t} \nabla \psi^*(\mathbf{r}, t) \right], \quad (2.21)$$

where  $\alpha$  is a proportionality constant depending on the choice of units. Since there is no energy transfer between true modes, the energy current density  $\mathbf{J} \cdot \hat{z}$  in the  $z$ -direction should be the same for  $z > z_{\alpha}$  and for  $z < z_{\alpha}$  for every mode  $|\psi_{\mathbf{k}}\rangle$  separately. In terms of the transmission and reflection, this leads to the requirement  $|\rho|^2 + |\tau|^2 = 1$ ; in terms of the T-matrix it reads

$$\text{Im } T(\mathbf{k}_{\parallel}, \omega) = -\frac{1}{2} \frac{|T(\mathbf{k}_{\parallel}, \omega)|^2}{k_z}. \quad (2.22)$$

This is the optical theorem for the plane scatterer. Note that it only holds for real  $k_z$ . The most general T-matrix satisfying the optical theorem has the form

$$T(\mathbf{k}_{\parallel}, \omega) = -[F^{-1}(\mathbf{k}_{\parallel}, \omega) - i/(2k_z)]^{-1}, \quad (2.23)$$

where the optical potential  $F(\mathbf{k}_{\parallel}, \omega)$  for the plane is a real function of its arguments. Furthermore, for planes that scatter isotropically, we have  $F(\mathbf{k}_{\parallel}, \omega) = F(k_{\parallel}, \omega)$ . The equations (2.20b) and (2.23) together show that a larger optical potential leads to a higher reflectivity of the plane.

### 2.3.2 N plane scatterers

Consider a system of  $N$  identical and parallel plane scatterers at arbitrary positions  $z_{\alpha}, z_{\beta}$ , and so on. At first, we do not assume that energy is conserved in the scattering process, so that the single-plane T-matrices need not be of the form (2.23). Like in the one-plane case, we want to know mode functions and Green functions of the  $N$ -plane system. This we do by calculating the T-matrix  $T^{(N)}(\mathbf{k}_{\parallel}, \omega)$ . Formally, we have as in the one-plane case that  $T^{(N)} = V(\mathbb{1} - g_0 V)^{-1}$ . The inverse is as yet unknown but according to Eq. (2.16) it can be written as an infinite series of products of single-plane T-matrices:

$$T^{(N)} = \sum_{\alpha} T_{\alpha} + \sum_{\beta \neq \alpha} \sum_{\alpha} T_{\beta} g_0 T_{\alpha} + \sum_{\gamma \neq \beta} \sum_{\beta \neq \alpha} \sum_{\alpha} T_{\gamma} g_0 T_{\beta} g_0 T_{\alpha} + \dots \quad (2.24)$$

Written out explicitly in the plane representation, the term of third order in the single-plane T-matrices in this series is

$$\frac{1}{(2\pi)^2} \sum_{\alpha} \sum_{\beta} \sum_{\gamma} \int d^2 \mathbf{k}_{\parallel} |\mathbf{k}_{\parallel}\rangle \langle \mathbf{k}_{\parallel}| T^3(\mathbf{k}_{\parallel}, \omega) g_0(\mathbf{k}_{\parallel}, z_{\alpha} - z_{\beta}, \omega) \times \\ (1 - \delta_{\alpha\beta}) g_0(\mathbf{k}_{\parallel}, z_{\beta} - z_{\gamma}, \omega) (1 - \delta_{\beta\gamma}) |z_{\alpha}\rangle \langle z_{\gamma}|. \quad (2.25)$$

Now define the matrix  $\mathcal{D}_{\alpha\beta}(\mathbf{k}_{\parallel}, \omega) \equiv g_0(\mathbf{k}_{\parallel}, z_{\alpha} - z_{\beta}, \omega)(1 - \delta_{\alpha\beta})$  and notice that the third-order term can be rewritten as

$$\frac{1}{(2\pi)^2} \sum_{\alpha} \sum_{\beta} \int d^2\mathbf{k}_{\parallel} |\mathbf{k}_{\parallel}\rangle \langle \mathbf{k}_{\parallel}| T(\mathbf{k}_{\parallel}, \omega) \{ [T(\mathbf{k}_{\parallel}, \omega) \mathcal{D}(\mathbf{k}_{\parallel}, \omega)]^2 \}_{\alpha\beta} |z_{\alpha}\rangle \langle z_{\beta}|. \quad (2.26)$$

So we can see the square of the matrix  $(T\mathcal{D})$  in the third-order terms. The fourth-order terms feature the cube of the matrix  $(T\mathcal{D})$ , and so on. This matrix structure enables us to sum the infinite series exactly: the T-matrix of  $N$  parallel and identical plane scatterers is given by

$$T^{(N)}(\omega) = \frac{1}{(2\pi)^2} \sum_{\alpha} \sum_{\beta} \int d^2\mathbf{k}_{\parallel} \sum_{\alpha, \beta} |\mathbf{k}_{\parallel}, z_{\alpha}\rangle T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) \langle z_{\beta}, \mathbf{k}_{\parallel}|, \quad (2.27)$$

where

$$T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) = T(\mathbf{k}_{\parallel}, \omega) [\mathbb{1} - T(\mathbf{k}_{\parallel}, \omega) \mathcal{D}(\mathbf{k}_{\parallel}, \omega)]_{\alpha\beta}^{-1}. \quad (2.28)$$

The problem of finding the T-matrix is now reduced to the much simpler problem of finding the inverse of the  $N \times N$  matrix  $\mathbf{T}^{(N)}$ . For arbitrary positions of the planes, the inversion must be done numerically. The matrix elements depend on the positions and effective thickness of the parallel planes, and on the frequency and in-plane wave vector of the light. Since equation (2.27) is valid for arbitrary positions of the plane scatterers, it could be the starting point for studying random scattering.

The important special case that will be studied here is the situation where the  $N$  planes are positioned at regular distances  $a$  from each other, at positions  $z_1 = a, z_2 = 2a, \dots, z_N = Na$ . We then have a three-dimensional optical Kronig-Penney model for a finite photonic crystal with periodic variation of the refractive index in one dimension. The regular distances between the planes enable us to do the inversion of the matrix in equation (2.28) analytically, as we shall discuss now.

Define  $\nu \equiv -2ik_z/T(\mathbf{k}_{\parallel}, \omega)$  and  $x \equiv \exp(ik_z a)$ . Then  $T_{\alpha\beta}^{(N)}$  in equation (2.28) can be rewritten as  $-2ik_z M_{\alpha\beta}^{-1}$  with

$$M_{kl} \equiv \nu \delta_{kl} + (1 - \delta_{kl}) x^{|k-l|} \quad (2.29)$$

We should like to know the inverse of this  $N \times N$  matrix  $\mathbf{M}$  for arbitrary  $N$  and in the appendix A this mathematical problem is solved. The result is

$$T_{\alpha\beta}^{(N)} = \frac{-2ik_z}{(2-\nu)x^2} \left\{ h(\alpha, \beta) - xh(\alpha+1, \beta) - xh(\alpha, \beta+1) + x^2h(\alpha+1, \beta+1) \right. \\ \left. + \frac{[xh(\alpha+1, 1) - h(\alpha, 1)][h(1, \beta) - xh(1, \beta+1)]}{1 + h(1, 1)} \right\} \quad (2.30)$$

This is the form of the T-matrix that is used in further calculations. (In appendix A it is shown how this expression can be rewritten such that certain symmetries of the matrix elements are more manifest.) The function  $h(\alpha, \beta) = h_N(\alpha, \beta)$  in Eq. (2.30) is defined as

$$h_N(\alpha, \beta) \equiv \frac{(2-\nu)x}{2(1-\nu)} \sum_{m=1}^N \left\{ \frac{\cos[(\alpha+\beta)m\pi/(N+1)] - \cos[(\alpha-\beta)m\pi/(N+1)]}{(N+1)(C - \cos[m\pi/(N+1)])} \right\}. \quad (2.31)$$

From the definition of  $h_N$  it follows that  $\mathbf{M}^{-1}$  is a symmetric matrix, as is  $\mathbf{M}$ . The matrix elements of  $\mathbf{T}^{(N)}$  strongly depend on the form of the constant  $C$  in equation (2.31) and for real  $k_z$  it is given by

$$C = \frac{2x^2 - \nu(1 + x^2)}{2x(1 - \nu)} = \cos(k_z a) + \sin(k_z a) \left[ \frac{2k_z \operatorname{Re}(T) + i(2k_z \operatorname{Im}(T) + |T|^2)}{4k_z^2 + |T|^2 + 4k_z \operatorname{Im}(T)} \right]. \quad (2.32)$$

Fortunately, the summation in equation (2.31) can be done exactly [73] so that we end up with a simpler expression for  $h_N$ :

$$h_N(\alpha, \beta) = \frac{(2 - \nu)x}{2(1 - \nu)} \left\{ \frac{\cos[(N + 1 - |\alpha - \beta|)Ka] - \cos[(N + 1 - (\alpha + \beta))Ka]}{\sin[Ka] \sin[(N + 1)Ka]} \right\}, \quad (2.33)$$

where the constant  $K = K(\mathbf{k}_{\parallel}, \omega)$  is defined by  $Ka = \arccos(C)$ . (The arccosine is a multiple-valued function but the function  $h_N$  is not; for numerical evaluation of  $h_N$  the Chebyshev polynomials are useful.) Evidently,  $K$  has dimension  $[\text{m}^{-1}]$  and in fact, it is equal to the Bloch wave vector of an infinite crystal of plane scatterers separated by a distance  $a$ . In general,  $K$  is complex. The important point that the Bloch wave vector that is normally associated with the infinite periodic potential also plays a role in the analysis of a finite crystal, was also stressed in [61]. Here the Bloch wave vector shows up in an analytical expression for the T-matrix of a finite crystal of arbitrary size  $N$ . No restrictions on the form of the single-plane T-matrix  $T(\mathbf{k}_{\parallel}, \omega)$  have been applied in the derivation.

Let us now assume that energy is conserved. This is the case when every single plane has a T-matrix that satisfies the optical theorem (2.23). Then it is evident from equation (2.32) that the constant  $C$  becomes real. Moreover, when the optical theorem holds, a simpler expression for  $C$  can be found by inserting for the single-plane T-matrix the form (2.23):

$$C = \cos(k_z a) - \left( \frac{F(\mathbf{k}_{\parallel}, \omega)}{2k_z} \right) \sin(k_z a). \quad (2.34)$$

One can see that without energy dissipation, the Bloch wave vector is either real (when  $-1 \leq C \leq 1$ ) or purely imaginary (otherwise). Imaginary Bloch wave vectors  $K(\mathbf{k}_{\parallel}, \omega)$  correspond to transmission stop band regions in  $(\mathbf{k}_{\parallel}, \omega)$ -space. This will be illustrated in section 2.4.1.

### 2.3.3 Model for the single plane scatterer

In the previous subsection, the T-matrix of the single plane scatterer was assumed to be known. Now we will derive a model for it, starting from a potential in the wave equation. Consider a dielectric slab of thickness  $d$  and dielectric function  $\varepsilon$ . In principle, the dielectric function is a frequency-dependent complex function [4], but here we neglect dispersion and loss altogether by choosing  $\varepsilon$  real and constant. Choose the  $z$ -axis such that it is perpendicular to the surfaces, defining the latter as the surfaces  $z = z_{\alpha}$  and  $z = z_{\alpha} + d$ . Then the scalar wave equation is

$$\left[ \frac{\varepsilon(\mathbf{r})}{c^2} \frac{\partial^2}{\partial t^2} - \nabla^2 \right] \psi(\mathbf{r}, t) = 0, \quad (2.35)$$

where  $\varepsilon(\mathbf{r}) = \varepsilon$  for  $z$  between  $z_\alpha$  and  $z_\alpha + d$ , and  $\varepsilon(\mathbf{r}) = 1$  elsewhere. Suppose now that the thickness of the slab is much smaller than the wavelengths of light we are interested in. Then we can model the slab as a plane scatterer. We define the product of slab thickness  $d$  and polarizability  $\varepsilon - 1$  as the effective thickness  $D_{\text{eff}}$  of the plane scatterer. We find the single-plane T-matrix by solving the associated equation for the Green function in frequency space:

$$[\nabla_r^2 + (\omega/c)^2] g(\mathbf{r}, \mathbf{r}', \omega) = \delta^3(\mathbf{r} - \mathbf{r}') + V(\mathbf{r}, \omega)g(\mathbf{r}, \mathbf{r}', \omega) \quad (2.36)$$

with the negative and frequency-dependent potential  $V(\mathbf{r}, \omega) = -(\omega/c)^2 D_{\text{eff}} \delta(z - z_\alpha)$ . It is easy to check that a solution of the Dyson-Schwinger equation (2.13) is a solution of the equation above, provided that in (2.13) we take the potential  $V(\omega) = -D_{\text{eff}}(\omega/c)^2 |z_\alpha\rangle\langle z_\alpha|$ . We shall now solve the Dyson-Schwinger equation, which in the mixed representation becomes

$$\begin{aligned} \langle \mathbf{k}_\parallel, z | g(\omega) | \mathbf{k}'_\parallel, z' \rangle &= (2\pi)^2 \delta(\mathbf{k}_\parallel - \mathbf{k}'_\parallel) g_0(\mathbf{k}_\parallel, z - z', \omega) \\ &+ g_0(\mathbf{k}_\parallel, z - z_\alpha, \omega) [-D_{\text{eff}}(\omega/c)^2] \langle \mathbf{k}_\parallel, z_\alpha | g(\omega) | \mathbf{k}'_\parallel, z' \rangle. \end{aligned} \quad (2.37)$$

If in this equation we put  $z$  equal to  $z_\alpha$ , then we can solve the equation for the Green function  $\langle \mathbf{k}_\parallel, z_\alpha | g(\omega) | \mathbf{k}'_\parallel, z' \rangle$ . If we insert this particular solution back into the equation (2.37) for general  $z$ , then we can also find the Green function for general  $z$ :  $\langle \mathbf{k}_\parallel, z | g(\omega) | \mathbf{k}'_\parallel, z' \rangle = (2\pi)^2 \delta(\mathbf{k}_\parallel - \mathbf{k}'_\parallel) g(\mathbf{k}_\parallel, z, z', \omega)$  with

$$g(\mathbf{k}_\parallel, z, z', \omega) = g_0(\mathbf{k}_\parallel, z - z', \omega) + g_0(\mathbf{k}_\parallel, z - z_\alpha, \omega) T(\mathbf{k}_\parallel, \omega) g_0(\mathbf{k}_\parallel, z_\alpha - z', \omega), \quad (2.38)$$

where the frequency- and wave vector-dependent T-matrix equals

$$T(\mathbf{k}_\parallel, \omega) = \frac{-D_{\text{eff}}(\omega/c)^2}{1 + g_0(\mathbf{k}_\parallel, 0, \omega) D_{\text{eff}}(\omega/c)^2} = -\left\{ [(\omega/c)^2 D_{\text{eff}}]^{-1} - i/(2k_z) \right\}^{-1}. \quad (2.39)$$

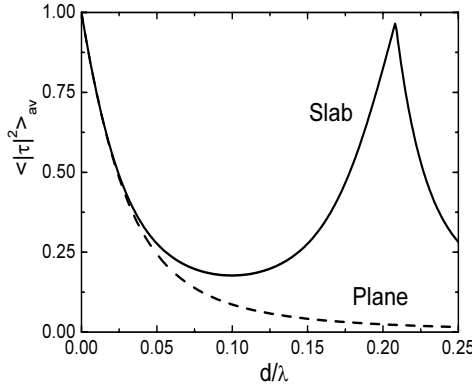
This T-matrix is of the form (2.23) and hence it satisfies the optical theorem (2.22). Note that  $T$  depends on  $k_\parallel$  rather than on  $\mathbf{k}_\parallel$ : the optical potential  $F(\mathbf{k}_\parallel, \omega)$  equals  $(\omega/c)^2 D_{\text{eff}}$  in this model, so that it depends only on frequency and not on the magnitude or direction of the wave vector of the incoming light.

A T-matrix of a point scatterer can only be defined after a careful regularization of the free-space Green function  $g_0(\mathbf{r}, \mathbf{r}', \omega)$  because the latter diverges in the limit  $|\mathbf{r} - \mathbf{r}'| \rightarrow 0$  [69]. Here, in the T-matrix for plane scatterers (2.39) it is the free-space Green function  $g_0(\mathbf{k}_\parallel, z - z', \omega)$  that appears in the limit of  $|z - z'| \rightarrow 0$ . Fortunately, there is no divergence so that we do not need a regularization procedure for plane scatterers.

Now that we have a one-parameter model for the T-matrix of a plane, we can use equations (2.20a) and (2.20b) to find the transmitted and reflected amplitudes  $\tau$  and  $\rho$ . The transmission amplitude is

$$\tau(\mathbf{k}_\parallel, \omega) = \frac{1}{1 - i D_{\text{eff}}(\omega/c)^2 / (2k_z)}. \quad (2.40)$$

Clearly, transmitted intensities range from one to zero when the effective thickness goes from zero to infinity.

**Figure 2.1:**

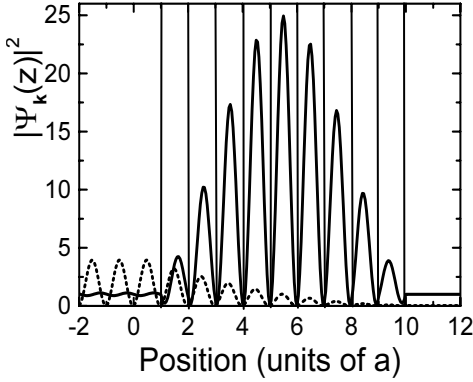
Solid line: angle-averaged transmission through a slab of refractive index  $n=2.6$  and thickness  $d$ , as a function  $d/\lambda$ . Dashed line: angle-averaged transmission through a plane scatterer with effective thickness  $D_{\text{eff}} = (n^2 - 1)d$ , as a function of  $d/\lambda$ .

In the rest of this section, we elaborate on the question how well slabs of finite thickness can be modeled as plane scatterers. We compare both cases by looking at the average transmission, with the average taken over  $4\pi$  solid angle. For the plane scatterer the average transmission can be calculated directly from equation (2.40), giving

$$\langle |\tau|^2 \rangle_{\text{av}} = 1 - (\pi D_{\text{eff}}/\lambda) \arctan [\lambda/(\pi D_{\text{eff}})]. \quad (2.41)$$

Light transmission through a finite slab is described by *Airy's formulae* [2] and the angle-average must be calculated numerically. Figure 2.1 shows the average transmission (for  $s$ -polarized light) of a  $\text{TiO}_2$  slab of thickness  $d$  in air as well as the average transmission of scalar waves through a plane scatterer with effective thickness  $D_{\text{eff}} = (n_{\text{TiO}_2}^2 - 1)d$ . The transmissions are plotted as a function of thickness  $d$  divided by the wavelength  $\lambda \equiv 2\pi c/\omega$ ; the refractive index  $n_{\text{TiO}_2}$  is 2.6. Figure 2.1 shows that slabs thinner than  $\lambda/20$  can be modeled adequately as plane scatterers with effective thickness  $D_{\text{eff}} = (n^2 - 1)d$ . Thicker slabs show resonances in the average transmission (due to interference effects) that the plane-scattering model does not describe. To give a concrete example, we consider a  $d = 26$  nm thick slab of  $\text{TiO}_2$ . If one constructs a Bragg mirror of such slabs alternating with air with a period of 260 nm, then evidently 10% of the Bragg mirror consists of  $\text{TiO}_2$ , which is also the typical number for three-dimensional photonic air-sphere crystals [33]. In normal direction the optical path length of the unit cell is 300 nm, so that the Bragg mirror has its first Bragg reflection at around 600 nm in that direction. At that wavelength we have  $d = 0.043\lambda$ , so that the single slab can indeed be described as a plane scatterer (and the Bragg mirror as a periodic array of plane scatterers). Our choice of  $D_{\text{eff}}$  proceeds only a bit different: we choose  $D_{\text{eff}} = 0.23\lambda_0$ , so that for wavelengths around  $\lambda_0 = 600$  nm the plane scatterer has the same average transmission of 32% as the slab. Furthermore, we choose the distance  $a$  between the planes to be 300 nm, so that the (blue edge of the) stop band in normal direction will be at  $\lambda_0 = 600$  nm.

In our scalar formalism we arrive at the same model for transmission and reflection of an infinitely thin plane as was found in Ref. [65] in another formalism, for  $s$ -polarized light. There, the authors start from a boundary condition analysis of the Maxwell fields around a dielectric slab in the limits  $\varepsilon \rightarrow \infty$  and  $d \rightarrow 0$  while keeping the product  $m = \varepsilon d$  constant. From our calculations and figure 2.1, it follows that it is actually the product

**Figure 2.2:**

Absolute values squared of two mode functions of a system of  $N = 10$  plane scatterers, as a function of the  $z$ -position in the structure. The dotted line corresponds to a mode with  $a/\lambda = 0.50$  and the solid line to a mode with  $a/\lambda = 0.51$ . Modes correspond to incoming plane waves from  $z = -\infty$  in the direction normal to the planes. The effective thickness of the plane scatterers is  $D_{\text{eff}} = 0.46a$ .

$D_{\text{eff}} = (\varepsilon - 1)d$  that should be kept constant while taking the limits. A comparison between plane scatterers for scalar and for vector waves will be made in chapter 3 of this thesis.

## 2.4 Optical modes

### 2.4.1 Propagating modes

With the T-matrix in hand in equation (2.30), we can use the solution (2.8) of the Lippmann-Schwinger equation to calculate the propagating modes of the finite photonic crystal. The propagating modes in our model are labeled by the wave vector and frequency of the corresponding incoming plane wave; actually they are related through  $(\omega/c)^2 = k^2$ . The propagating mode  $|\psi_{\mathbf{k}}(\omega)\rangle$  corresponding to an  $N$ -plane crystal is given by

$$\psi_{\mathbf{k}}(\mathbf{r}, \omega) = e^{i\mathbf{k} \cdot \mathbf{r}} - \frac{i}{2k_z} e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}} \sum_{\alpha, \beta=1}^N e^{ik_z|z-z_{\alpha}|} T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) e^{ik_z z_{\beta}}. \quad (2.42)$$

If we put  $N = 1$  in this equation, then we find back the mode function (2.20a) for the single plane. In figure 2.2 the absolute values squared of the mode functions are plotted for two modes propagating normal to a crystal of  $N = 10$  planes. (We choose the normal direction, but any direction is possible.) As is clear from the figure, the slight frequency difference of the modes has an enormous influence on the mode profile in the crystal: the mode with  $a/\lambda = 0.5$  is almost completely reflected, as the maximum amplitude of almost 4 of the almost standing wave at the left hand side of the structure indicates; the mode with  $a/\lambda = 0.51$  has an amplitude that grows inside the structure and is almost completely transmitted. As can be seen from equation (2.42) and from figure 2.2, the absolute value squared of  $\psi_{\mathbf{k}}(\mathbf{r}, \omega)$  is a constant for  $z > Na$ . This is the (intensity) transmission  $|\tau|^2$  through the  $N$ -plane system. Since there are no losses, reflection and transmission add up to one. In figure 2.3 the reflection is plotted for light incoming in the normal direction, as a function of frequency. The prominent features are the stop bands, the frequency intervals for which light cannot enter the crystal in this direction. The mode

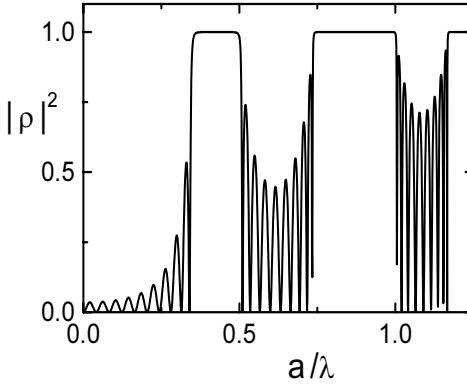


**Figure 2.3:** Reflectivity  $|\rho|^2$  of normally incident waves by a crystal of  $N = 10$  plane scatterers separated by a distance  $a$ , as a function of  $a/\lambda$ .

with  $a/\lambda = 0.5$  of figure 2.2 falls just inside the first stop band in figure 2.3, whereas the other mode corresponds to the first reflection minimum at the blue side of this first stop band. The stop bands correspond to frequencies for which the Bloch wave vector (proportional to the cosine of equation (2.23)) is a complex number. In between the stop bands, the Bloch wave vector is real and there one finds the reflection maxima and minima which are characteristic for periodic media. In figure 2.3 higher-order stop bands become broader. This is because the optical potential  $D_{\text{eff}}(\omega/c)^2$  rapidly grows with frequency since we do not take the frequency dispersion of  $D_{\text{eff}}$  into account. Of course, one could improve on this by taking dispersion into account and writing  $D_{\text{eff}} = [\varepsilon(\omega) - 1] d$ , but the optical theorem (2.22) only holds as long as the complex part of  $\varepsilon(\omega)$  can be neglected.

Transmission and mode profiles of propagating modes in layered systems usually are calculated with the transfer matrix technique [2, 55]. Here we showed that for a crystal of plane scatterers one can alternatively use the T-matrix formalism. For presentation purposes we chose  $N = 10$ , but we also considered mode profiles and reflection for two hundred planes and more planes do not form a problem: the T-matrix elements of  $N$  planes are known analytically and the calculation of the mode function boils down to a simple summation over the  $N^2$  matrix elements  $T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega)$ .

### 2.4.2 Guided modes

In the previous section we found a continuum of propagating modes with real wave vector components in all three directions. The crystals of plane scatterers could also support a finite number of guided modes that propagate along the planes with in-plane wave vectors  $k_{\parallel} > \omega/c$  so that  $k_z = i\kappa$  and  $k_{\parallel}^2 - \kappa^2 = (\omega/c)^2$ . The imaginary wave vector in the  $z$ -direction makes that guided modes decay exponentially outside the scatterer: unlike propagating modes, guided modes do not correspond to an incident wave. It is the dispersion relations of the guided modes that we concentrate on here; the contribution of the guided modes to the local density of states is discussed in section 2.5.

As was said in section 2.2, in general the bound states of a scattering system can be found as the solutions of the Lippmann-Schwinger equation in the absence of an incident wave [Eq. (2.10)]. In particular, a guided mode  $|\psi_{\kappa}\rangle$  of the crystal of plane scatterers is

bound in the  $z$ -direction and it satisfies

$$\psi_\kappa(z, \omega) = \int_{-\infty}^{+\infty} dz_1 g_0(\mathbf{k}_\parallel, z - z_1, \omega) V(z_1, \omega) \psi_\kappa(z_1, \omega), \quad (2.43)$$

with the potential  $V(z, \omega) = -F \sum_{n=1}^N \delta(z - na)$  and  $F = D_{\text{eff}}(\omega/c)^2$  and the free-space Green function as defined in (2.19). Given a frequency  $\omega$  and the number of planes  $N$ , this equation will have a finite number of solutions. In order to find them, define the  $N$ -dimensional vector  $\Psi$  with components  $\Psi_j = \psi_\kappa(z_j)$  in which the  $z_j$  are the plane positions. Using (2.43), the following homogeneous linear system of equations can be derived:

$$\sum_{\beta=1}^N [\delta_{\alpha\beta} + g_0(k_\parallel, z_\alpha - z_\beta, \omega) F] \Psi_\beta = 0 \quad \text{for } \alpha = 1, 2, \dots, N. \quad (2.44)$$

Nontrivial solutions of (2.44) only exist if the determinant of the coefficient matrix vanishes. Now this coefficient matrix can be rewritten as  $-F(\mathbf{T}^{(N)})_{\alpha\beta}^{-1}$  with the use of the general definition of the T-matrix,  $T \equiv V(\mathbb{1} - g_0 V)^{-1}$ . So the guided modes can be found as the poles of the T-matrix. This is the direct connection between the guided modes and the T-matrix.

The guided modes of a single plane are easy to find. For  $N = 1$ , the T-matrix  $T_{11}^{(1)}(\mathbf{k}_\parallel, \omega)$  of equation (2.30) reduces to  $T(k_\parallel, \omega)$  as in equation (2.39). The one-plane T-matrix has a pole  $\kappa_1^{(1)} = (\omega/c)^2 D_{\text{eff}}/2$ . This is the dispersion relation for the guided mode of a single plane. (One could equivalently present the dispersion relation as  $\omega$  versus  $k_\parallel$ , but here and in the following, we choose to present  $\omega$  versus  $\kappa$ .) The dispersion relation shows that for every frequency, a single plane has one and only one guided mode. The same single guided mode for a single plane was found in [65] using another formalism; the case of finite  $N > 1$  was not considered there.

Why does a single plane scatterer have only one guided mode? Dielectric slabs of finite thickness can support more than one guided mode (depending on its thickness  $d$  and dielectric constant  $\varepsilon$ ). These guided modes other than the first one are orthogonal to this first one. Their mode functions have their extreme values inside the slab and change sign at least once inside the slab. In the limit of an infinitely thin slab, the mode functions remain continuous, so that the maxima, the minima, and the zeroes should coincide on the plane. Therefore, a plane scatterer can support at most one guided mode.

How many guided modes exist in a crystal of  $N$  plane scatterers? We have just derived that they occur when the determinant  $\det[(\mathbf{T}^{(N)})^{-1}]$  is equal to zero. With the definition of the matrix  $\mathbf{M}$  in equation (2.29) this condition simplifies to  $\det[\mathbf{M}(k_\parallel, \omega)] = 0$ . A formula for this determinant is given in equation (A.14) of appendix A, for an arbitrary number of planes. The result is that guided modes correspond to solutions of  $1 + h_N(1, 1) = 0$ , with  $h_N(\alpha, \beta)$  given in equation (2.33). We assume that the planes do not absorb light and use the form (2.23) for the single-plane T-matrix. Then the condition for guides modes to exist becomes

$$[2\kappa + F(\mathbf{k}_\parallel, \omega)] e^{-\kappa a} \frac{\sin(NKa)}{\sin[(N+1)Ka]} = 2\kappa. \quad (2.45)$$

This single equation determines the number of guided modes that can exist at a certain frequency for an arbitrary system size  $N$ . The frequency-dependence of the equation is in the Bloch wave vector  $K$  and in the optical potential  $F$ .

A new guided mode appears at a certain frequency with  $\kappa = 0$  and when the frequency becomes higher, this mode becomes more localized to the structure so that  $\kappa$  increases. Thus, we can count guided modes at their point of first appearance, at  $\kappa = 0$ . There we find that the condition (2.45) simplifies to  $\sin(NKa) = 0$ . Now for wave vectors  $k_z = i\kappa$ , the Bloch wave vector satisfies

$$\cos(Ka) = \cosh(\kappa a) - [F/(2\kappa)] \sinh(\kappa a), \quad (2.46)$$

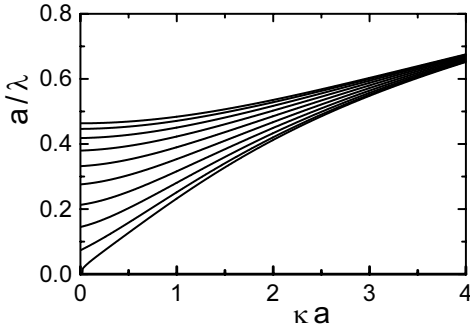
so that with  $F = D_{\text{eff}}(\omega/c)^2$  we find guided modes with frequencies

$$\omega_m(\kappa = 0) = \sqrt{2c^2/(aD_{\text{eff}})} \sqrt{1 - \cos(m\pi/N)} \quad (m = 0, 1, \dots, N-1). \quad (2.47)$$

So it turns out that for a systems of  $N$  plane scatterers at a fixed frequency, there are at most  $N$  guided modes. All  $N$  guided modes emerge in the frequency interval  $0 \leq \omega \leq 2c/\sqrt{aD_{\text{eff}}}$ . An increase in the effective thickness of the planes makes this frequency interval smaller, because at the same frequency the modes have larger  $\kappa$  and are more localized, just like was analyzed for the single plane above. Or the other way round, for fixed  $\kappa$  the frequency of a mode becomes lower when the effective thickness increases. In the limit  $N \rightarrow \infty$  a band of guided modes is formed. The case of an infinite number of planes was considered in [65] and there indeed the band at  $\kappa = 0$  is given by the frequency interval given above.

Figure 2.4 shows the dispersion of the modes  $\omega_m(\kappa)$  as a function of  $\kappa$ , in units of the plane separation  $a$ , for  $N = 10$ . Actually, the frequencies are plotted as  $a/\lambda$ , and all guided modes are indeed seen to appear at  $\kappa = 0$  when  $0 \leq a/\lambda \leq (1/\pi)\sqrt{a/D_{\text{eff}}} = 0.47$ . The figure illustrates that when the product  $\kappa a$  is much bigger than one, the planes guide light independently so that the  $N$  modes all have frequencies near the one-plane frequency  $\sqrt{2\kappa/D_{\text{eff}}}$ . For smaller  $\kappa a$ , the  $N$  planes interact more strongly and the frequencies of the modes become more separated. So going from right to left in the figure, we go from  $N$  independent single-plane guided modes to  $N$  solutions of a genuine  $N$ -plane problem. The physical way to traverse the plot is of course by changing the frequency: at frequencies just below  $2c/\sqrt{aD_{\text{eff}}}$  there are both well-delocalized and well-localized modes. At higher frequencies, all guided modes are localized, whereas at lower frequencies the inverse decay lengths  $(\kappa)^{-1}$  are larger so that only delocalized modes are to be expected. The localization of the guided modes will be illustrated in section 2.5.

Now let us compare figure 2.4 with figure 2.3 which showed the stop bands for light falling on the planes in the normal direction. The first stop band in normal direction occurs for light with wavelengths  $0.35 \leq a/\lambda \leq 0.50$ . Light propagation normal to the planes is strongly suppressed. In this same frequency interval, the four last guided modes of the structure appear. More generally, for our choice of  $D_{\text{eff}}$ , all the interesting behavior of the guided modes appears around the first stop band for light propagating in the normal direction. Even when the plane crystal blocks light coming from almost all directions, then light of this same frequency can be guided inside the structure. When  $D_{\text{eff}}$  is decreased, the stop bands become narrower, whereas the frequency interval in which the guided modes appear becomes wider.

**Figure 2.4:**

Dispersion relations of the guided modes for a ten-plane system. The  $\kappa$  on the horizontal axis equals  $-ik_z$ . The effective thickness of the plane scatterers is  $D_{\text{eff}} = 0.46a$ .

## 2.5 Local optical density of states

In inhomogeneous dielectrics, the spontaneous-emission rate of a radiating atom depends on its position and is proportional to a quantity that is often called the local density of states (LDOS). In quantum optics, many quantum states of light can be associated with what in this thesis we call an optical ‘mode’, so that the words ‘state’ and ‘mode’ have a different meaning. Actually, with “density of states” it is meant “density of modes” and indeed the latter term is sometimes used [60, 61]. But we stick to the familiar term LDOS and assume that it will not confuse the reader. The LDOS is a function of position and frequency and will be denoted by  $\rho(\mathbf{r}, \omega)$ . (This  $\rho$  is different from the reflection amplitude of section 2.3.3.)

For scalar waves in three dimensions,  $\rho(\mathbf{r}, \omega)$  is  $-2\omega/(\pi c^2)$  times the imaginary part of the Green function  $g(\mathbf{r}, \mathbf{r}, \omega)$ . In free space, all modes are radiative (or propagating) modes and  $\rho_0(\mathbf{r}, \omega) = \omega^2/(2\pi^2 c^3)$ , independent of position. In dielectric structures, the LDOS is the sum of the local density of radiative modes  $\rho_r(\mathbf{r}, \omega)$  and of the local density of guided modes  $\rho_g(\mathbf{r}, \omega)$ , which we will also call the radiative and guided LDOS, respectively. Spontaneous emission (or in this chapter: its scalar analogy) can occur into both types of modes, the physical difference between the two cases being that light emitted into a propagating mode eventually can be detected in the far field, whereas light emitted into the guided modes does not leave the structure.

First we need to know the Green function of the crystal of plane scatterers. Often a Green function is given and calculated in terms of its mode decomposition, but here we proceed differently. The same T-matrix (2.30) that gave the propagating and guided modes when inserted in the Lippmann-Schwinger equation, also gives the Green function when inserted in the Dyson-Schwinger equation (2.13):

$$\begin{aligned}
 g(\mathbf{k}_{\parallel}, z, z', \omega) &= g_0(\mathbf{k}_{\parallel}, z - z', \omega) \\
 &+ \sum_{\alpha, \beta=1}^N g_0(\mathbf{k}_{\parallel}, z - z_{\alpha}, \omega) T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) g_0(\mathbf{k}_{\parallel}, z_{\beta} - z', \omega).
 \end{aligned} \tag{2.48}$$

This is the Green function in the mixed representation and in order to find the LDOS we must transform it to real space using a two-dimensional inverse Fourier transformation and then take the imaginary part. The crystal is translation invariant in the  $x$ - and  $y$ -directions

so that the LDOS depends on the  $z$ -position only. Moreover, in our model (2.39) the individual planes scatter light isotropically so that also  $T_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) = T_{\alpha\beta}^{(N)}(k_{\parallel}, \omega)$ . Then the integral over directions  $\hat{k}_{\parallel}$  is trivial; the integral over the magnitude  $k_{\parallel}$  is split into a propagating and a guided part, as we present below.

The radiative LDOS is given by

$$\begin{aligned} \rho_r(z, \omega) &= \rho_0(z, \omega) \\ &- \frac{\omega}{\pi^2 c^2} \text{Im} \int_0^{\omega/c} dk_{\parallel} k_{\parallel} \sum_{\alpha, \beta=1}^N g_0(k_{\parallel}, z - z_{\alpha}, \omega) T_{\alpha\beta}^{(N)} g_0(k_{\parallel}, z_{\beta} - z, \omega). \end{aligned} \quad (2.49)$$

The integral is taken over the magnitude of the in-plane wave vectors  $k_{\parallel}$  with values between 0 and  $\omega/c$ , so that all wave vectors contributing to the radiative LDOS are real.

The remaining part of the inverse Fourier transform gives the guided LDOS:

$$\rho_g(z, \omega) = -\frac{\omega}{\pi^2 c^2} \text{Im} \int_{\omega/c}^{\infty} dk_{\parallel} k_{\parallel} \sum_{\alpha, \beta=1}^N g_0(k_{\parallel}, z - z_{\alpha}, \omega) T_{\alpha\beta}^{(N)}(k_{\parallel}, \omega) g_0(k_{\parallel}, z_{\beta} - z, \omega). \quad (2.50)$$

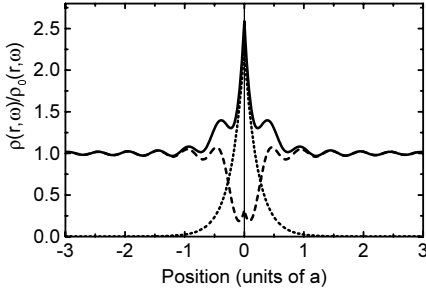
Here  $k_{\parallel} > \omega/c$ , so that the integrand in (2.50) is real except at the poles  $k_{\parallel}(\omega)$  of the T-matrix elements. Now the T-matrix elements are written in equation (2.30) in terms of the functions  $h_N$ , but it can be shown that the poles of the latter do not correspond to poles of the  $T_{\alpha\beta}^{(N)}$ . The T-matrix elements only have poles when  $1 + h_N(1, 1) = 0$ , corresponding to the guided modes that were found in section 2.4.2. The right-hand side of the equality (2.50) therefore simplifies into a sum over residues of the integrand at the guided-mode poles. The infinitesimally small positive imaginary parts of the poles ensure that  $\rho_g(z, \omega) \geq 0$ .

Now we discuss what the LDOS actually looks like in a system of plane scatterers. First consider  $\rho_r$  for a single plane located at  $z = 0$ . The T-matrix is given in equation (2.39). Exactly at the position of the plane, the relative radiative LDOS  $\rho_r(0, \omega)/\rho_0(0, \omega)$  turns out to be equal to the average transmission  $\langle |\tau|^2 \rangle_{\text{av}}$  through the plane (equation (2.41)). As expected, highly reflecting planes affect  $\rho_r$  more strongly. Figure 2.5 shows the local densities in the neighborhood of the plane. (Although for  $N = 1$  the distance between the planes  $a$  is not defined, we can choose a distance  $a$  and scale the plot such that  $D_{\text{eff}} = 0.46a$  and  $a/\lambda = 0.5$ .) At  $z = 0$  we indeed have  $\rho_r/\rho_0 = 0.32$ , equal to the average transmission of the plane. Away from the plane,  $\rho_r$  oscillates back to  $\rho_0$ .

Figure 2.5 also shows the guided LDOS for the single plane at  $z = 0$ . With the pole of the T-matrix located at  $\kappa_1^{(1)} = D_{\text{eff}}(\omega/c)^2/2 + i\eta$ , the guided LDOS (2.50) in this case is

$$\rho_g(\mathbf{r}, \omega) = \left( \frac{-2\omega}{\pi c^2} \right) \left( \frac{-\kappa_1^{(1)}}{4} \right) \exp(-2\kappa_1^{(1)}|z|). \quad (2.51)$$

So for  $N = 1$ , the guided LDOS falls off exponentially as a function of distance from the plane, with a decay length of  $[\kappa_1^{(1)}(\omega)]^{-1}$ . At the position of the plane the relative guided LDOS  $\rho_g/\rho_0$  is equal to  $\pi^2 D_{\text{eff}}/\lambda$ , which is equal to 2.27 for the values of the parameters

**Figure 2.5:**

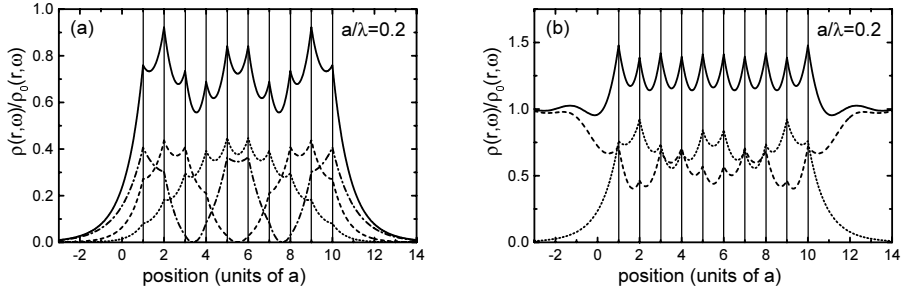
Local densities of states for a single plane, relative to the free-space value, for  $a/\lambda = 0.5$  and  $D_{\text{eff}} = 0.46a$ . Both the radiative (dashed line) and the guided LDOS (dotted line) are shown, as well as their sum (solid line).

of figure 2.5. So we know how at the position of the plane  $\rho_g$  goes up and  $\rho_r$  goes down when the effective thickness is increased.

For larger systems, for  $N = 10$  say, the guided modes can either be localized to individual planes or be delocalized over the entire structure. At least, this was anticipated in section 2.4.2 when discussing the dispersion relations of the guided modes. Now we can check this. Figures 2.6(a-b) show the radiative and guided LDOS as well as their sum, as a function of position in and around the structure, for a frequency such that  $a/\lambda = 0.2$ . Again, the densities were divided by the free space value for a good comparison. Local densities of individual modes are plotted in figure 2.6(a). According to figure 2.4 there are three guided modes at this frequency, with  $\kappa_1^{(10)}a = 0.84$ ,  $\kappa_2^{(10)}a = 0.77$  and  $\kappa_3^{(10)}a = 0.48$ , respectively. From figure 2.6(a) one can learn that each of these three guided modes is delocalized over all the planes and also that every next guided mode has one extra node in its mode density. The guided LDOS corresponds to the solid line in figure 2.6(a); it is the sum of the contributions of the three modes and it does not behave periodically inside the crystal.

Note that  $\rho_g(\mathbf{r}, \omega)$  divided by the free-space value varies around 0.7 inside the structure. The guided modes are therefore an important channel for spontaneous emission. This is also illustrated by figure 2.6(b), where for the same frequency the guided LDOS, the radiative LDOS, as well as their sum are shown. The frequency is well below the first stop band in any direction (see figure 2.3). As is clear from figure 2.6(b), inside the structure  $\rho_r(z, \omega)$  is smaller than  $\rho_g(z, \omega)$ . Notice also that the sum  $\rho_r + \rho_g$  shows more periodicity in the structure than  $\rho_r$  or  $\rho_g$  separately. Outside of the structure the radiative LDOS climbs back to the free space value, showing damped oscillations; the guided LDOS drops down to zero. The decay looks exponential with a decay length of about  $\lambda/4$ . In fact, the decay is multi-exponential because all three guided modes decay with their own decay length. Moreover, according to equation (2.50) the exponential decay does not start at the outer planes only.

Figure 2.7(a) also shows local densities of states, but for a higher frequency such that  $a/\lambda = 0.5$ . This frequency is interesting, because it is the location of the blue edge of the first stop band in normal direction, as can be seen in figure 2.3. Light that makes an angle  $\theta$  with the normal has a first stop band that is shifted to higher frequencies, according to Bragg's law  $2a \cos \theta = \lambda$ . At  $a/\lambda = 0.5$ , stop bands of many directions

**Figure 2.6:**

Local densities of states for a crystal of plane scatterers with  $N = 10$ ; densities are divided by the free-space density. Figure (a) shows the guided-mode LDOS (solid line) at  $a/\lambda = 0.2$ . It is the sum of contributions from three guided modes: the first (dotted line) without nodes, the second one (dashed) with one node and the third guided mode (dot-dashed) with two nodes. Figure (b) shows at the same frequency the guided LDOS (dotted line), the radiative LDOS (dashed) as well as the total LDOS (solid line).

will overlap. Therefore it is to be expected that  $\rho_r(\mathbf{r}, \omega)$  is much lower than in free space. Figure 2.7(a) shows precisely this: the  $\rho_r(\mathbf{r}, \omega)$  rapidly drops down to almost zero inside the plane crystal. Actually, what we have in figure 2.7(a) is an omni-directional mirror [59]. This can be checked by varying the angle of the incoming light in equation (2.42) (not shown): for all angles ranging from 0 to 90 degrees, the first stop bands overlap at  $a/\lambda = 0.5$ . Figure 2.7(a) suggests a working definition of a finite omni-directional mirror: *A periodic structure is an omni-directional mirror at a frequency  $\omega$  when the maximum value of  $\rho_r(z, \omega)/\rho_0(z, \omega)$  in the middle unit cell is smaller than some previously agreed-upon value  $\sigma \ll 1$ .* The parameter  $\sigma$  denotes the maximum negligible amount of radiative LDOS that one accepts inside a finite omni-directional mirror. For example, the structure of figure 2.7(a) only counts as an omni-directional mirror when we accept a  $\sigma$  of 0.013 or larger. Only for infinite structures can one find omni-directional mirrors with  $\sigma = 0$ . Although the number  $N$  of unit cells does not explicitly enter this definition, it is clear that increasing  $N$  can help to turn a structure into an omni-directional mirror.

Figure 2.7(a) also shows that the guided LDOS  $\rho_g(\mathbf{r}, \omega)$  does not go to zero in the omni-directional mirror. At  $a/\lambda = 0.5$ , the maximum number of ten guided modes have appeared in the ten-plane crystal. Since inside the structure the total LDOS is almost exclusively made up of  $\rho_g(\mathbf{r}, \omega)$ , spontaneous emission at this frequency occurs into one of these guided modes with almost 100 percent efficiency. Conversely, the extraction efficiency  $\rho_r/(\rho_g + \rho_r)$  of light spontaneously emitted inside the crystal will be almost zero at  $a/\lambda = 0.5$ .

Finally, in figure 2.7(b) the frequency is increased such that  $a/\lambda = 0.6$ , above the first stop band in the normal direction (see figure 2.3). Then incident light in and around the normal direction can enter the structure again and therefore  $\rho_r(\mathbf{r}, \omega)$  is nonzero inside

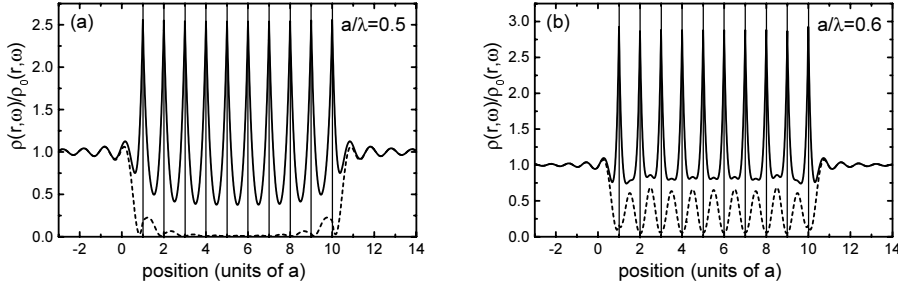


**Figure 2.7:**

Radiative (dashed) and total (solid) LDOS of a ten-plane crystal, for  $a/\lambda = 0.5$  in figure (a) and  $a/\lambda = 0.6$  in figure (b). Not shown in both is the guided LDOS, which is the difference between total and radiative LDOS.

the crystal, as the figure shows. For this frequency, the structure is not an omni-directional mirror for any reasonable tolerance parameter  $\sigma$ . The combined density of the ten guided modes has become even more localized to the planes, which is the only thing that happens to  $\rho_g$  when increasing the frequency even more. Interestingly, in between the planes the radiative LDOS is small where the guided LDOS is large, and vice versa, so that the variation in the total LDOS is small. Near the planes, however, the total LDOS is much larger than the free-space value, mainly due to  $\rho_g$ .

In general, the radiative LDOS can not be increased much beyond the free-space value, at least for the effective thickness  $D_{\text{eff}} = 0.46a$  that we consider. This is somewhat surprising since individual propagating modes can be strongly resonant inside the structure (see figure 2.2). But  $\rho_r(z, \omega)$  is an integral (2.49) over all directions, some of resonant and others of decaying nature. As it turns out, this does not add up to form a large increase. Even if the fraction of light spontaneously emitted into a certain direction can be dramatically increased compared to the emission in vacuum, the total emission rate into radiative modes is not increased much.

A last remark on the mirror symmetry of the LDOS in figure 2.5. The LDOS should of course be symmetrical in  $z = (N + 1)a/2$ . In our formalism, this corresponds to a symmetry in the T-matrix elements:  $T_{\alpha, \beta}^{(N)} = T_{N+1-\alpha, N+1-\beta}^{(N)}$ . In particular, this symmetry should also be found in the residues of  $T_{\alpha, \beta}^{(N)}$  at the poles  $\kappa_m^{(N)}(\omega)$  corresponding to the guided modes. We could use the symmetry in the plots as a check on our calculations, because the plots were not made symmetrical ‘by hand’. Note that from Eq. (2.30) it is not obvious whether the T-matrix elements have the required symmetry at all; in appendix A it is shown that the matrix elements can be rewritten in a form in which the symmetry is manifest.

## 2.6 Summary and conclusions

In this chapter we have used the plane scatterer as our work horse and discussed its range of validity. We put it to use in the T-matrix formalism of multiple-scattering theory. The T-matrix of  $N$  identical and parallel planes located at arbitrary positions was found as the inverse of a known  $N \times N$  matrix, in equation (2.28); using the trick as discussed in appendix A, in equation (2.30) an explicit expression for the T-matrix elements of a crystal of regularly-spaced plane scatterers was given.

These analytical results enabled us to find relatively easily both the radiative modes (in equation (2.42)) and the guided modes (defined by equation (2.45)) of a crystal consisting of an arbitrary but finite number of planes. Not only individual modes, but also the complete Green function could be found (equation (2.48)). From the Green function, the local density of states can be calculated, which is the sum of a radiative and a guided LDOS. The sum of the two determines the local spontaneous-emission rate and the relative contribution of the radiative to the total LDOS gives you the extraction efficiency of spontaneously emitted light. Clearly, in order to make a connection with experiments, it is important to know the Green function of finite crystals. The knowledge of the Green function and the LDOS is also needed for the interpretation of images recorded with a scanning near-field optical microscope [74–76]. Here especially the LDOS near the surface of the crystal is important.

We chose to present results for  $N = 10$ . The ten-plane crystal was shown to exhibit stop bands. Around the frequencies of the first stop band in normal direction, guided modes appear as delocalized over the whole crystal whereas for higher frequencies they localize to the individual planes. Around  $a/\lambda = 0.5$  the crystal is an omni-directional mirror. We argued to consider a finite periodic structure as an omni-directional mirror if the maximal value of the radiative LDOS in its innermost unit cell is smaller than some previously agreed-upon finite value. The LDOS of the crystal of planes is a well-behaved function of position and frequency. The variations inside a unit cell can be large. Evidently, the finite translation symmetry of the LDOS is broken for the finite crystals, especially near the surfaces. The total LDOS shows more translation symmetry than the radiative or guided LDOS separately, which can be seen as a competition between both types of modes with different outcomes when going from one unit cell to another.



## Chapter 3

# Spontaneous emission of vector waves in crystals of plane scatterers

*The concept of a plane scatterer that was developed in the previous chapter for scalar waves is generalized to Maxwell vector waves. T-matrices can be defined after a Green function regularization. Optical modes and Green functions are determined and differences with scalar waves are stressed. The theory is used to calculate position- and orientation-dependent spontaneous-emission rates and radiative line shifts.*

### 3.1 Multiple-scattering theory for vector waves

In section 2.1 a general introduction and motivation to study optical properties of crystals of plane scatterers was given. Since chapter 2 dealt with scalar waves only and since light is a vector wave, it is essential to study how the concept of a plane scatterer can be generalized to vector waves. This is the subject of the present chapter.

The general multiple-scattering theory as introduced in section 2.2 for scalar waves described by the Helmholtz equation, can be generalized to vector waves that satisfy Maxwell's equations. The notation inevitably becomes more involved: scalar quantities such as frequency  $\omega$  or components of vectors or matrices such as the wave vector component  $k_z$  will be written in standard font; vectors will be written in bold standard font, for example the electric field " $\mathbf{E}_{\mathbf{k}\sigma}$ "; finally, matrices or tensors such as the free-space Green function " $\mathbf{G}_0$ " will also be written bold but in a *sans serif* font.

The wave equation for the electric field  $\mathbf{E}_0(\mathbf{r}, \omega)$  in vacuum is

$$\{(\omega/c)^2 \mathbf{I} \cdot -\nabla \times \nabla \times \} \mathbf{E}_0(\mathbf{r}, \omega) = 0. \quad (3.1)$$

The symbol  $\mathbf{I}$  denotes the identity operator in three-dimensional space. The solutions of Eq. (3.1) are plane waves with wave vector  $\mathbf{k}$  and polarization direction normal to  $\mathbf{k}$ .

With the wave equation (3.1) the free-space Green tensor (or *dyadic* Green function) is associated that satisfies

$$\{(\omega/c)^2 \mathbf{I} \cdot -\nabla \times \nabla \times\} \mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) = \delta^3(\mathbf{r} - \mathbf{r}') \mathbf{I}. \quad (3.2)$$

Now it will be convenient to consider Eq. (3.1) as the real-space representation of an abstract tensor operator  $\mathbf{L}(\omega)$  acting on the vector field  $\mathbf{E}_0(\omega)$ , so that (3.1) in abstract notation becomes

$$\mathbf{L}(\omega) \cdot \mathbf{E}_0(\omega) = 0. \quad (3.3)$$

In the same notation the dyadic Green function  $\mathbf{G}_0(\omega)$  satisfies

$$\mathbf{L}(\omega) \cdot \mathbf{G}_0(\omega) = \mathbf{1} \otimes \mathbf{I} \quad (3.4)$$

The identity operator in real space we denote by  $\mathbf{1}$  and it has the property  $\langle \mathbf{r} | \mathbf{1} | \mathbf{r}' \rangle = \delta^3(\mathbf{r} - \mathbf{r}')$ ; confusion with the unit tensor  $\mathbf{I}$  should not arise; the  $\otimes$  denotes the tensor product.

In the presence of an inhomogeneous dispersive linear dielectric, the wave equation for the electric field is modified as follows:

$$\begin{aligned} \{(\omega/c)^2 \mathbf{I} \cdot -\nabla \times \nabla \times\} \mathbf{E}(\mathbf{r}, \omega) &= -[(\varepsilon(\mathbf{r}, \omega) - 1)(\omega/c)^2] \mathbf{E}(\mathbf{r}, \omega) \\ &\equiv \mathbf{V}(\mathbf{r}, \omega) \cdot \mathbf{E}(\mathbf{r}, \omega), \end{aligned} \quad (3.5)$$

where in the last equality the frequency-dependent optical potential  $\mathbf{V}$  was defined in terms of the dielectric function  $\varepsilon(\mathbf{r}, \omega)$ . The electric field  $\mathbf{E}_{\text{in}}(\omega)$  is modified into  $\mathbf{E}(\omega)$ , and the two fields are related through the Lippmann-Schwinger equation

$$\mathbf{E}(\omega) = \mathbf{E}_{\text{in}}(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{E}(\omega). \quad (3.6)$$

The field  $\mathbf{E}(\omega)$  that satisfies Eq. (3.6) is also a solution of Eq. (3.5). The solution of Eq. (3.6) can be found iteratively in higher and higher orders of the optical potential  $\mathbf{V}$ :

$$\mathbf{E} = \mathbf{E}_{\text{in}} + \mathbf{G}_0 \cdot \mathbf{V} \cdot \mathbf{E}_{\text{in}} + \mathbf{G}_0 \cdot \mathbf{V} \cdot \mathbf{G}_0 \cdot \mathbf{V} \cdot \mathbf{E}_{\text{in}} + \dots \quad (3.7)$$

So we have a multiple-scattering series for the electric field in terms of the potential, both when the potential consists of many scatterers and when it models just a single scatterer. A (dyadic) T-matrix can be defined that sums up how the electric field depends on incoming field and scatterer:

$$\mathbf{E}(\omega) = \mathbf{E}_{\text{in}}(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{E}_{\text{in}}(\omega). \quad (3.8)$$

This is a formal definition of the T-matrix as a  $3 \times 3$  tensor, and by combining Eqs. (3.7) and (3.8), the formal solution for the T-matrix is

$$\mathbf{T}(\omega) = \mathbf{V}(\omega) \cdot [\mathbf{I} - \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega)]^{-1}. \quad (3.9)$$

The scattering problem is solved exactly once the T-matrix is known.

As we saw for scalar waves, there may exist electric field modes that are bound to the scatterer. Such bound modes are solutions of the Lippmann-Schwinger equation in the absence of an incident field:

$$\mathbf{E}(\omega) = \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{E}(\omega). \quad (3.10)$$

With the help of the formal definition (3.9) of the T matrix, we can rewrite this homogeneous equation as

$$\mathbf{V}(\omega) \cdot \mathbf{T}^{-1}(\omega) \cdot \mathbf{E}(\omega) = 0. \quad (3.11)$$

This equation shows that nontrivial bound solutions of the electric field will correspond to the poles of the T-matrix. So the T-matrix not only solves the scattering problem for incident fields but also contains all information about bound modes.

In the presence of the dielectric the Green function also changes from  $\mathbf{G}_0$  to  $\mathbf{G}$  and the latter satisfies the following equation

$$[\mathbf{L}(\omega) - \mathbf{V}(\omega)] \cdot \mathbf{G}(\omega) = \mathbf{1} \otimes \mathbf{I}, \quad (3.12)$$

The solution for the Green function analogous to Eq. (3.6) for the electric field is the three-dimensional Dyson-Schwinger equation

$$\mathbf{G}(\omega) = \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{V}(\omega) \cdot \mathbf{G}(\omega). \quad (3.13)$$

It can be verified that a solution of (3.13) also is a solution of equation (3.12). The problem how to find such a solution is solved once the T-matrix (3.9) is determined, because an iteration of Eq. (3.13) analogous to the series expansion (3.7) for the electric field shows that the Green function can also be expressed in terms of the T-matrix:

$$\mathbf{G}(\omega) = \mathbf{G}_0(\omega) + \mathbf{G}_0(\omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{G}_0(\omega). \quad (3.14)$$

The equations (3.13) and (3.14) also hold when the total potential  $\mathbf{V}(\omega)$  is a sum of single-scatterer potentials  $\mathbf{V}_\alpha(\omega)$ . By iterating one finds that the total T-matrix for an arbitrary number  $N$  of these scatterers is

$$\mathbf{T}^{(N)} = \sum_{\alpha} \mathbf{V}_{\alpha} + \sum_{\beta} \sum_{\alpha} \mathbf{V}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{V}_{\alpha} + \sum_{\gamma} \sum_{\beta} \sum_{\alpha} \mathbf{V}_{\gamma} \cdot \mathbf{G}_0 \cdot \mathbf{V}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{V}_{\alpha} + \dots \quad (3.15)$$

Again, it is much more convenient to do the expansion in terms of the single-plane T-matrices:

$$\mathbf{T}^{(N)} = \sum_{\alpha} \mathbf{T}_{\alpha} + \sum_{\beta \neq \alpha} \sum_{\alpha} \mathbf{T}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\alpha} + \sum_{\gamma \neq \beta} \sum_{\beta \neq \alpha} \sum_{\alpha} \mathbf{T}_{\gamma} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\alpha} + \dots \quad (3.16)$$

The equations of multiple-scattering theory for vector waves were introduced, in the notation that will be used throughout this chapter. The analogies with and differences from scalar waves can be found when comparing with section 2.2.

## 3.2 Plane scatterers for vector waves

The elements of the scattering theory are the potential  $\mathbf{V}$ , the free-space Green function  $\mathbf{G}_0$ , the T-matrix  $\mathbf{T}$ , and the incoming electric field  $\mathbf{E}_{\text{in}}$ . Here we will determine the specific form that these elements take for dielectrics that can be described as a collection of parallel planes. First the free-space Green function is determined, followed by the single-plane T-matrix and finally the T-matrix for an arbitrary number of planes.

### 3.2.1 Dyadic Green function in plane representation

A solution for the dyadic Green function can be found in three-dimensional Fourier space and by translational invariance of free space we have  $\langle \mathbf{k} | \mathbf{G}_0(\omega) | \mathbf{k}' \rangle = (2\pi)^3 \delta^3(\mathbf{k} - \mathbf{k}') \mathbf{G}_0(\mathbf{k}, \omega)$ . The Green function  $\mathbf{G}_0(\mathbf{k}, \omega)$  that we thus defined satisfies

$$\{[(\omega/c)^2 - k^2] \mathbf{I} + k^2 \hat{\mathbf{k}} \hat{\mathbf{k}}\} \cdot \mathbf{G}_0(\mathbf{k}, \omega) = \mathbf{I}. \quad (3.17)$$

Here,  $\hat{\mathbf{k}}$  denotes a unit vector in the direction of the wave vector  $\mathbf{k}$ . Equation (3.17) is a  $3 \times 3$  matrix equation whose representation diagonalizes in the polarization basis  $\{\hat{\mathbf{k}}, \hat{\sigma}_1, \hat{\sigma}_2\}$  with the longitudinal direction  $\hat{\mathbf{k}}$  and two orthogonal transverse directions  $\hat{\sigma}_{1,2}$ . Note that this representation is co-rotating with the wave vector. The solution of (3.17) is

$$\begin{aligned} G_0^{jj}(\mathbf{k}, \omega) &= \frac{1}{(\omega/c)^2 - k^2} \\ G_0^{\hat{\mathbf{k}}\hat{\mathbf{k}}}(\mathbf{k}, \omega) &= (c/\omega)^2, \end{aligned} \quad (3.18)$$

where  $j$  denotes  $\sigma_1$  or  $\sigma_2$ . All six non-diagonal elements of the Green tensor are zero in this representation.

The spatial Fourier representation is not what we need. As in chapter 2, it is convenient to work in the “plane representation”: in two-dimensional Fourier space in the directions parallel to the planes and in real space in the  $\hat{\mathbf{z}}$ -direction perpendicular to the planes. For the polarization representation we choose the orthonormal basis  $\{\hat{\mathbf{s}}_{\mathbf{k}}, \hat{\mathbf{v}}_{\mathbf{k}}, \hat{\mathbf{z}}\}$ . Here,  $\hat{\mathbf{z}}$  is the unit vector in the  $z$ -direction;  $\hat{\mathbf{v}}_{\mathbf{k}}$  is the unit vector in the direction of the projection of the wave vector  $\mathbf{k}$  on the plane, so that the wave vector  $\mathbf{k}$  has a wave vector component  $k_{\parallel}$  in the  $\hat{\mathbf{v}}_{\mathbf{k}}$ -direction and its full representation is  $(0, k_{\parallel}, k_z)$ ; the  $\hat{\mathbf{s}}_{\mathbf{k}}$ -polarization direction is orthogonal to the optical plane that is spanned by the other two basis vectors. Then the operator  $\mathbf{L}(\omega)$  has the form  $\langle \mathbf{k}_{\parallel}, z | \mathbf{L}(\omega) | \mathbf{k}'_{\parallel}, z' \rangle = (2\pi)^2 \delta^2(\mathbf{k}_{\parallel} - \mathbf{k}'_{\parallel}) \delta(z - z') \mathbf{L}(\mathbf{k}_{\parallel}, z, \omega)$  and the operator  $\mathbf{L}(\mathbf{k}_{\parallel}, z, \omega)$  has the matrix representation

$$\mathbf{L}(\mathbf{k}_{\parallel}, z, \omega) = \begin{pmatrix} (\omega/c)^2 - k_{\parallel}^2 + \partial_z^2 & 0 & 0 \\ 0 & (\omega/c)^2 + \partial_z^2 & -ik_{\parallel} \partial_z \\ 0 & -ik_{\parallel} \partial_z & (\omega/c)^2 - k_{\parallel}^2 \end{pmatrix}. \quad (3.19)$$

The wave vector  $\mathbf{k}_{\parallel}$  is a two-dimensional in-plane wave vector. The Green function in the same representation becomes  $\langle \mathbf{k}_{\parallel}, z | \mathbf{G}_0(\omega) | \mathbf{k}'_{\parallel}, z' \rangle = (2\pi)^2 \delta^2(\mathbf{k}_{\parallel} - \mathbf{k}'_{\parallel}) \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega)$ , and the matrix equation (3.17) becomes a system of differential equations in the plane representation:

$$\mathbf{L}(\mathbf{k}_{\parallel}, z, \omega) \begin{pmatrix} G_0^{ss} & G_0^{sv} & G_0^{sz} \\ G_0^{vs} & G_0^{vv} & G_0^{vz} \\ G_0^{zs} & G_0^{zv} & G_0^{zz} \end{pmatrix} = \delta(z - z') \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (3.20)$$

The  $G_0^{pq}$  are the components of  $\mathbf{G}_0$  and the arguments  $(\mathbf{k}_{\parallel}, z, z', \omega)$  were dropped for brevity. By choosing this representation, the matrix elements of  $\mathbf{G}_0$  only depend on the magnitude and not on the orientation of  $\mathbf{k}_{\parallel}$ . Evidently, all components involving an  $s$ -label

are zero except for the  $ss$ -component that has an equation that is uncoupled from the rest. Actually,  $G_0^{ss}$  satisfies the differential equation for the Green function of scalar waves that we encountered in the previous chapter. So we have

$$G_0^{ss}(\mathbf{k}_{\parallel}, z, z', \omega) = \frac{e^{ik_z|z-z'|}}{2ik_z}, \quad (3.21)$$

where  $k_z$  is not a new independent variable different from  $\mathbf{k}_{\parallel}$ , but rather an abbreviation for  $\sqrt{(\omega/c)^2 - k_{\parallel}^2}$ . The remaining differential equations are coupled and can also be solved. It turns out that  $G_0^{vv}$  is proportional to  $G_0^{ss}$  that is already known. Once  $G_0^{vv}$  is known,  $G_0^{zv}$  immediately follows. When solving for  $G_0^{vz}$ , the equation for  $G_0^{vz}$  is satisfied by the solution for component  $G_0^{zv}$  that we just found, so that the two these components are equal. The last nonzero component  $G_0^{zz}$  can then be expressed in terms of the others and the results are

$$\begin{aligned} G_0^{vv}(\mathbf{k}_{\parallel}, z, z', \omega) &= \frac{k_z^2}{(\omega/c)^2} \frac{e^{ik_z|z-z'|}}{2ik_z} \\ G_0^{vz}(\mathbf{k}_{\parallel}, z, z', \omega) &= -\frac{k_{\parallel}k_z}{(\omega/c)^2} \frac{e^{ik_z|z-z'|}}{2ik_z} \text{sign}(z - z') \\ G_0^{zv}(\mathbf{k}_{\parallel}, z, z', \omega) &= G_0^{vz}(\mathbf{k}_{\parallel}, z, z', \omega) \\ G_0^{zz}(\mathbf{k}_{\parallel}, z, z', \omega) &= \frac{k_{\parallel}^2}{(\omega/c)^2} \frac{e^{ik_z|z-z'|}}{2ik_z} + (c/\omega)^2 \delta(z - z'). \end{aligned} \quad (3.22)$$

The above method of solving differential equations does not give a clue as to what the value of the sign-function for  $z$  equal to  $z'$  should be. It appears that there is another way to obtain the results above, one that does give the value of  $\text{sign}(0)$ , namely by using the Fourier relation between the Green functions in the two representations:

$$\mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk_z \mathbf{G}_0(\mathbf{k}_{\parallel}, k_z, \omega) e^{ik_z(z-z')}. \quad (3.23)$$

The integration can only be performed in a representation that does not co-rotate with  $k_z$ . The basis of Eq. (3.18) is not adequate, but again the basis  $\{\hat{\mathbf{s}}_{\mathbf{k}}, \hat{\mathbf{v}}_{\mathbf{k}}, \hat{\mathbf{z}}\}$  suits well. Again we find the Green function components of Eq. (3.22), with the additional information that integrands asymmetric in the variable  $k_z$  lead to a sign-function that is zero when  $z$  equals  $z'$ . The free-space dyadic Green function in the plane representation is now determined.

### 3.2.2 Attempt to define a T-matrix

The T-matrix of a plane scatterer for vector waves can be found by solving the appropriate Lippmann-Schwinger (LS) equation (3.6), just as was done in the previous chapter for scalar waves. A plane wave incident from  $z = -\infty$  with wave vector  $\mathbf{k}$  and arbitrary amplitude  $E_0$  and transverse polarization vector  $\boldsymbol{\sigma}_{\mathbf{k}} = (\sigma_s, \sigma_v, \sigma_z)$  is scattered by a plane at  $z = z_{\alpha}$ . Because of the symmetry in the in-plane directions, it is convenient to choose the plane representation for the LS equation. In terms of the Dirac-notation, the electric

field is a “ket” and the plane representation is found by taking the inner product of equation (3.6) for the electric field with the “bra”  $\langle \mathbf{k}_{\parallel}, z |$ , and by inserting the unit operator

$$\frac{1}{(2\pi)^2} \int d^2 \mathbf{k}'_{\parallel} dz' |\mathbf{k}'_{\parallel}, z'\rangle \langle \mathbf{k}'_{\parallel}, z'| \quad (3.24)$$

at the positions of the dots in the representation-independent equation (3.6). The incident field takes the form  $\mathbf{E}_{\mathbf{k}\sigma, \text{in}}(\mathbf{k}_{\parallel}, z, \omega) = E_0 \sigma_{\mathbf{k}} \exp(ik_z z)$ . The solution of the LS equation corresponding to this incident field is the field  $\mathbf{E}_{\mathbf{k}\sigma}(\omega)$  and note that it is labelled by the wave vector and polarization of the incoming field from which it originates. The plane itself is described by the optical potential  $\mathbf{V}(z, \omega) = -[\varepsilon(z, \omega) - 1](\omega/c)^2 \mathbf{I} \equiv V(\omega)\delta(z - z_{\alpha})\mathbf{I}$ . Then the Lippmann-Schwinger equation in the mixed representation becomes

$$\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z, \omega) = E_0 \sigma_{\mathbf{k}} e^{ik_z z} + \int_{-\infty}^{+\infty} dz' \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega) \cdot \mathbf{V}(z', \omega) \cdot \mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z', \omega). \quad (3.25)$$

The integral is very simple because the planes are assumed to be infinitely thin, and we get

$$\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z, \omega) - V(\omega) \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z_{\alpha}, \omega) = E_0 \sigma_{\mathbf{k}} e^{ik_z z}. \quad (3.26)$$

The analogous equation for scalar waves could be solved at this point by putting the position  $z$  in this equation equal to  $z_{\alpha}$ , solving for  $\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z_{\alpha}, \omega)$  and putting back this result in the above equation to obtain an expression for  $\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z_{\alpha}, \omega)$ . However, unlike the scalar Green function  $g_0$ , the Green tensor  $\mathbf{G}_0$  is not defined when the positions  $z$  and  $z_{\alpha}$  are identical, because of the delta function in the component  $G_0^{zz}$  [Eq. (3.22)]. Therefore it is impossible with this Green tensor to set up a scattering theory for vector waves scattered by infinitely thin planes. In the next section we propose a regularization of the Green function in order to overcome the problem encountered above. We could just neglect the delta function, as is sometimes done in other calculations [77], but as we shall see later in subsection 3.2.4, this procedure does not give the correct results in our case.

### 3.2.3 Regularization of the Green function

When studying multiple scattering of a wave by objects much smaller than the wavelength, the assumption that the scatterers are infinitely thin or small can simplify the analysis considerably. Often it is the only route to analytical results for a truly multiply-scattering system. Thus, spatially extended physical objects are modelled as mathematical objects with zero volume. But this simplification comes at a price. One may run into the kind of problem encountered in the previous section, that a Green function does not have a finite value precisely at the position of a zero-volume-scatterer. This technical complication can be remedied by a procedure called “regularization”.

When modelling scatterers not as infinitely thin planes but as mathematical points, the above problem of unphysical infinities shows up both for scalar and for vector waves. For point scatterers the problem has been studied extensively and several regularization schemes have been proposed (see [69] and references therein). In a regularization procedure usually some cutoff parameter is introduced that modifies the behavior of Green

functions at distances much smaller than optical wavelengths and infinities are thus removed.

After a successful regularization, two situations can present themselves. In the first situation, the optical properties are not affected by the procedure, in the sense that the value of the cutoff parameter can be chosen infinitely large in the final stage. In this first case, the mathematical problem of an infinity could be solved with mathematics and the scattering theory is called “renormalizable”.

In the second situation, the cutoff parameter can not be sent to infinity. It should then be possible to express the finite cutoff parameter in terms of physical quantities. But then it also works the other way around: physical properties depend on the cutoff parameter. In this second situation, the theory is not renormalizable and the physical parameters together with the cutoff parameter form an overdetermined set. The latter situation was found for point scatterers for vector waves [69]. A T-matrix for a point scatterer could only be defined when two regularization parameters were introduced, namely a longitudinal and a transverse cutoff parameter. The longitudinal cutoff is related to a static quantity, namely the natural size of the scatterer; the transverse cutoff can be expressed either in terms of dynamic quantities (for example: resonance frequencies) or static quantities, the latter choice depending on the physical scatterer that is modelled as a point. So much for general remarks about Green function regularization.

In the rest of this subsection, we regularize the dyadic Green function with components as in Eq. (3.22). We propose the same regularization procedure as was done for point scattering, namely a high-momentum cutoff in three-dimensional Fourier space: instead of the free-space Green function  $\mathbf{G}_0(\mathbf{k}, \omega)$  of Eq. (3.18) we use the regularized free-space Green function  $\tilde{\mathbf{G}}_0(\mathbf{k}, \omega)$ , and define the latter in terms of the former as

$$\tilde{\mathbf{G}}_0(\mathbf{k}, \omega) = \frac{\Lambda^2}{\Lambda^2 + k^2} \mathbf{G}_0(\mathbf{k}, \omega). \quad (3.27)$$

Here, the cutoff momentum  $\Lambda$  is assumed to be much larger than any optical momentum, so that at optical wavelengths  $\tilde{\mathbf{G}}_0 \simeq \mathbf{G}_0$ . Now we are interested what the effect of this cutoff will be in the plane representation. For the unregularized Green function, we showed that the plane representation could be found either by solving differential equations (3.20) or by an inverse Fourier transform (3.23). The latter method is the simplest for finding the regularized Green function in the plane representation, and we find:

$$\begin{aligned} \tilde{G}_0^{ss}(\mathbf{k}_{\parallel}, z, z_1, \omega) &= \frac{\Lambda^2}{\Lambda^2 + (\omega/c)^2} \left( \frac{e^{ik_z|z-z_1|}}{2ik_z} + \frac{e^{-\Lambda_{\parallel}|z-z_1|}}{2\Lambda_{\parallel}} \right) \\ \tilde{G}_0^{vv}(\mathbf{k}_{\parallel}, z, z_1, \omega) &= \frac{k_z^2 c^2}{\omega^2} \tilde{G}_0^{ss}(\mathbf{k}_{\parallel}, z, z_1, \omega) \\ \tilde{G}_0^{vz}(\mathbf{k}_{\parallel}, z, z_1, \omega) &= -\frac{k_{\parallel} k_z c^2}{\omega^2} \text{sign}(z - z_1) \tilde{G}_0^{ss}(\mathbf{k}_{\parallel}, z, z_1, \omega) \\ \tilde{G}_0^{zv}(\mathbf{k}_{\parallel}, z, z_1, \omega) &= \tilde{G}_0^{vz}(\mathbf{k}_{\parallel}, z, z_1, \omega) \\ \tilde{G}_0^{zz}(\mathbf{k}_{\parallel}, z, z_1, \omega) &= \frac{k_{\parallel}^2 c^2}{\omega^2} \tilde{G}_0^{ss}(\mathbf{k}_{\parallel}, z, z_1, \omega) + \frac{\Lambda^2 e^{-\Lambda_{\parallel}|z-z_1|}}{2(\omega/c)^2 \Lambda_{\parallel}}. \end{aligned} \quad (3.28)$$

In these equations,  $\Lambda_{\parallel}$  is short-hand notation for  $\sqrt{\Lambda^2 + k_{\parallel}^2}$ . (Again, the sign-function is zero when its argument is.) All components of the regularized Green tensor consist of two parts: an oscillating and a decaying part, as a function of  $|z - z_1|$ . The decay is very fast because the decay length  $\Lambda^{-1}$  is much smaller than the optical wavelength  $\lambda = 2\pi c/\omega$ . For  $\lambda|z - z_1| \gtrsim 1$  and  $\Lambda \gg \lambda$ , the regularized Green function approaches the unregularized one. If one would take the limit of  $\Lambda \rightarrow \infty$ , then all the components in (3.28) approach the unregularized components of equation (3.22), and in particular the limit of the last term in  $\mathbf{G}_0^{zz}$  gives the delta function that made the regularization procedure necessary. However, we keep  $\Lambda$  finite for the moment. Unlike  $G_0^{zz}$ , the component  $\tilde{G}_0^{zz}$  does not have a term proportional to  $\delta(z - z_1)$ , but it has a term that grows with  $\Lambda$  instead. This enables us to set up a theory of scattering by vector waves from plane scatterers.

### 3.2.4 T-matrix of a plane for vector waves

In subsection 3.2.2, we stumbled on a problem to define the T-matrix of a plane for vector waves. After completing the regularization of the Green function in subsection 3.2.3, we now go back to the Lippmann-Schwinger equation (3.25) and replace the Green function by the regularized one:

$$\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z, \omega) = E_0 \sigma_{\mathbf{k}} e^{ik_z z} + \int_{-\infty}^{+\infty} dz' \tilde{\mathbf{G}}_0(\mathbf{k}_{\parallel}, z, z', \omega) \cdot \mathbf{V}(z', \omega) \cdot \mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z', \omega). \quad (3.29)$$

Now take  $z = z_{\alpha}$  in equation (3.29) and write it out in components:

$$\begin{pmatrix} E_{\mathbf{k}\sigma}^s \\ E_{\mathbf{k}\sigma}^v \\ E_{\mathbf{k}\sigma}^z \end{pmatrix} = E_0 \begin{pmatrix} \sigma_s \\ \sigma_v \\ \sigma_z \end{pmatrix} e^{ik_z z_{\alpha}} + V(\omega) \begin{pmatrix} \tilde{G}_0^{ss} & 0 & 0 \\ 0 & \tilde{G}_0^{vv} & 0 \\ 0 & 0 & \tilde{G}_0^{zz} \end{pmatrix} \begin{pmatrix} E_{\mathbf{k}\sigma}^s \\ E_{\mathbf{k}\sigma}^v \\ E_{\mathbf{k}\sigma}^z \end{pmatrix}. \quad (3.30)$$

Here,  $\tilde{G}_0^{ss} = \tilde{G}_0^{ss}(\mathbf{k}_{\parallel}, z_{\alpha}, z_{\alpha}, \omega)$  and similarly for the other components. The off-diagonal elements of the Green tensor are all zero when the position  $z$  is equal to  $z_{\alpha}$ . Hence the equation can be solved for every component separately. If we insert this result into the Lippmann-Schwinger equation for general  $z$ , we find

$$\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z, \omega) = \mathbf{E}_{\mathbf{k}\sigma, \text{in}}(\mathbf{k}_{\parallel}, z, \omega) + \tilde{\mathbf{G}}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \tilde{\mathbf{T}}(\mathbf{k}_{\parallel}, \omega) \cdot \mathbf{E}_{\mathbf{k}\sigma, \text{in}}(\mathbf{k}_{\parallel}, z_{\alpha}, \omega), \quad (3.31)$$

where the T-matrix for scattering from a plane by arbitrarily polarized light is given by

$$\tilde{\mathbf{T}}(\mathbf{k}_{\parallel}, \omega) = \begin{pmatrix} \frac{V(\omega)}{1 - V(\omega)\tilde{G}_0^{ss}} & 0 & 0 \\ 0 & \frac{V(\omega)}{1 - V(\omega)\tilde{G}_0^{vv}} & 0 \\ 0 & 0 & \frac{V(\omega)}{1 - V(\omega)\tilde{G}_0^{zz}} \end{pmatrix}. \quad (3.32)$$

This T-matrix is what we were looking for. Now that it is known we can calculate transmission and reflection and local densities of states as well as other physical quantities.

The scattering of the  $s$ -polarization component of the light can be considered independently from the  $\hat{\mathbf{v}}$  and  $\hat{\mathbf{z}}$  directions, according to Eq. (3.31). It can be verified with

Eqs. (3.28, 3.32) that since  $\Lambda \gg (\omega/c)$ , the matrix component  $\tilde{T}^{ss}$  for all practical purposes is equal to the T-matrix for scalar waves, and the same holds for the Green tensor component  $\tilde{G}_0^{ss}$ : the regularization was not necessary for  $s$ -polarized light and fortunately it does not affect the scattering properties of  $s$ -polarized light either.

The need for regularization did show up in the description of scattering of  $p$ -polarized light and it is interesting to analyze how the cutoff influences the scattering properties. Assume that  $p$ -polarized light is impinging on a plane, with amplitude  $E_0$ , wave vector  $\mathbf{k}$ , and the polarization state

$$\hat{\sigma} = \hat{p} \equiv (k_z/k)\hat{\mathbf{v}}_{\mathbf{k}} - (k_{\parallel}/k)\hat{\mathbf{z}}. \quad (3.33)$$

Written out explicitly, the incoming field is

$$\mathbf{E}_{\mathbf{k}\sigma, \text{in}}(\mathbf{k}_{\parallel}, z, \omega) = \begin{pmatrix} E_{\mathbf{k}\sigma, \text{in}}^s(\mathbf{k}_{\parallel}, z, \omega) \\ E_{\mathbf{k}\sigma, \text{in}}^v(\mathbf{k}_{\parallel}, z, \omega) \\ E_{\mathbf{k}\sigma, \text{in}}^z(\mathbf{k}_{\parallel}, z, \omega) \end{pmatrix} = E_0 \begin{pmatrix} 0 \\ k_z/k \\ -k_{\parallel}/k \end{pmatrix} e^{ik_z z}. \quad (3.34)$$

The planes do not mix the  $s$  and  $p$ -polarization so that the incoming-plus-scattered field does not have an  $s$ -component either. For the nonzero components we find:

$$\begin{pmatrix} E^v(z) \\ E^z(z) \end{pmatrix} = \begin{pmatrix} E_{\text{in}}^v(z) + \tilde{G}_0^{vv}(z, z_{\alpha})\tilde{T}^{vv}E_{\text{in}}^v(z_{\alpha}) + \tilde{G}_0^{vz}(z, z_{\alpha})\tilde{T}^{zz}E_{\text{in}}^z(z_{\alpha}) \\ E_{\text{in}}^z(z) + \tilde{G}_0^{zv}(z, z_{\alpha})\tilde{T}^{vv}E_{\text{in}}^v(z_{\alpha}) + \tilde{G}_0^{zz}(z, z_{\alpha})\tilde{T}^{zz}E_{\text{in}}^z(z_{\alpha}) \end{pmatrix}, \quad (3.35)$$

where the labels  $(\mathbf{k}_{\parallel}, \sigma)$  were dropped for readability. Now for distances far enough from the plane so that  $\Lambda|z - z_{\alpha}| \gg 1$ , the term  $\tilde{G}_0^{vz}\tilde{T}^{zz}$  falls off as  $\Lambda^{-1} \exp(-\Lambda|z - z_{\alpha}|)$  and  $\tilde{G}_0^{zz}\tilde{T}^{zz}$  as  $\exp(-\Lambda|z - z_{\alpha}|)$ , so that for optical purposes these terms can be neglected. For finite very large  $\Lambda$  we arrive at the following effective description:

$$\begin{pmatrix} E^s(z) - E_{\text{in}}^s(z) \\ E^v(z) - E_{\text{in}}^v(z) \\ E^z(z) - E_{\text{in}}^z(z) \end{pmatrix} = \begin{pmatrix} G_0^{ss} & 0 & 0 \\ 0 & G_0^{vv} & G_0^{vz} \\ 0 & G_0^{zv} & G_0^{zz} \end{pmatrix} \begin{pmatrix} T^{ss} & 0 & 0 \\ 0 & T^{vv} & 0 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_{\text{in}}^s(z_{\alpha}) \\ E_{\text{in}}^v(z_{\alpha}) \\ E_{\text{in}}^z(z_{\alpha}) \end{pmatrix}, \quad (3.36)$$

where the  $ss$ -component of the T-matrix is equal to  $V(\omega)[1 - V(\omega)G_0^{ss}]^{-1}$ , and analogously for the  $vv$ -component. The Green functions have arguments  $(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega)$ . In this effective description, - where the T-matrix is denoted by  $\mathbf{T}$  rather than  $\tilde{\mathbf{T}}$  - the cutoff parameter  $\Lambda$  does not occur anymore. The cutoff was necessary in order to set up a scattering theory and it shows up in the elements of the scattering theory such as the T-matrix (3.32) and the regularized Green function (3.28). But it does not show up in the electric field and precisely this enables us to arrive at the effective description. Note also that in the effective description the value of  $G_0^{zz}$  has become irrelevant.

Can the delta function in  $G_0^{zz}$  (see Eq. (3.22)) just be left out of the theory, as is sometimes done in other work [77], and produce the same T-matrix? The answer is No. Leaving out the delta function in the Lippmann-Schwinger equation (3.25) will result in a nonzero  $T^{zz}$ , in contrast with Eq. (3.36). Furthermore, the  $T$  matrix would be such that the transmitted part of an incoming wave would not be parallel to this incoming wave, which is unphysical. The conclusion is that a regularization of the Green function was necessary, even when in the end the scattering theory is renormalizable, in the sense described in section 3.2.3.

### 3.2.5 Transmission and energy conservation

The transmission of light through the plane can be found by choosing  $z > z_\alpha$  in Eq. (3.36). The transmitted wave can be expressed in terms of the incoming wave as  $\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_\parallel, z, \omega) = \boldsymbol{\tau}(\mathbf{k}_\parallel, \omega) \cdot \mathbf{E}_{\mathbf{k}\sigma, \text{in}}(\mathbf{k}_\parallel, z, \omega)$ , with the transmission matrix

$$\boldsymbol{\tau}(\mathbf{k}_\parallel, \omega) = \begin{pmatrix} \tau_{ss}(\mathbf{k}_\parallel, \omega) & 0 & 0 \\ 0 & \tau_{vv}(\mathbf{k}_\parallel, \omega) & 0 \\ 0 & \tau_{zv}(\mathbf{k}_\parallel, \omega) & 1 \end{pmatrix} \quad (3.37)$$

which has nonzero elements  $\tau_{jj}(\mathbf{k}_\parallel, \omega) = [1 - V(\omega)G_0^{jj}(k_\parallel, z_\alpha, z_\alpha, \omega)]^{-1}$  for  $j = s, v$ . Furthermore,  $\tau_{zv}(\mathbf{k}_\parallel, \omega) = G^{zv}(k_\parallel, z_\alpha, z_\alpha, \omega)T^{vv}(k_\parallel, \omega)$ . With the help of Eqs. (3.22, 3.34, 3.36) one can find that both for  $s$ -polarized and for  $p$ -polarized light, the transmitted electric field is a polarization-dependent scalar times the incoming electric field vector. In other words, after transmission the direction is unchanged but the amplitude can be different from the incoming wave.

Now we can ask which forms the T-matrix can take such that energy is conserved in the scattering process. This has been analyzed for scalar waves before and since  $s$ -waves map on scalar waves, we know the optical theorem for the  $ss$ -component of the T matrix:

$$\text{Im } T^{ss}(\mathbf{k}_\parallel, \omega) = -\frac{1}{2} \frac{|T^{ss}(\mathbf{k}_\parallel, \omega)|^2}{k_z}. \quad (3.38)$$

The most general T-matrix satisfying this requirement has the form

$$T^{ss}(\mathbf{k}_\parallel, \omega) = -[F_s^{-1}(\mathbf{k}_\parallel, \omega) - i/(2k_z)]^{-1}, \quad (3.39)$$

with the optical potential  $F_s(\mathbf{k}_\parallel, \omega)$  being a real-valued function.

Energy conservation of light implies a conservation of the  $\hat{z}$ -component of the Poynting vector before and after the plane. For reflection and transmission of  $p$ -polarized light, only the matrix element  $T^{vv}$  is important and again we are interested in the form that this matrix element can take. An incoming plane  $p$ -wave with polarization (3.33) gives the electric field (3.36) and with a Maxwell equation the accompanying magnetic field can also be found. The Poynting vector is then [45]

$$\mathbf{S}(\mathbf{r}, t) = \frac{c}{2\pi\mu_0} \text{Re} [\mathbf{E}^*(\mathbf{r}, t) \times \mathbf{B}(\mathbf{r}, t)]. \quad (3.40)$$

First transform the equations to frequency space. At the side of the plane where light is reflected ( $z < z_\alpha$ ), the Poynting vector is then proportional to  $1 - (k_z c/\omega)^2 |T^{vv}|^2/4$ . This should be equal to  $|1 - ik_z(c/\omega)^2 T^{vv}/2|^2$ , the expression that one finds for the other side. From energy conservation one can obtain the optical theorem for the scattering of  $p$ -polarized light by a plane:

$$\text{Im } T^{vv}(\mathbf{k}_\parallel, \omega) = \frac{-k_z}{2(\omega/c)^2} |T^{vv}(\mathbf{k}_\parallel, \omega)|^2. \quad (3.41)$$

This expression differs from the optical theorem for  $s$ -polarized light. Also, the most general solution of the optical theorem is different:

$$T^{vv}(\mathbf{k}_{\parallel}, \omega) = - \left[ F_p^{-1}(\mathbf{k}_{\parallel}, \omega) - \frac{ik_z}{2(\omega/c)^2} \right]^{-1}, \quad (3.42)$$

where the optical potential  $F_p(\mathbf{k}_{\parallel}, \omega)$  is real.

### 3.2.6 T-matrix for $N$ planes

The Green function and the T-matrix of a single plane are known now, and with this a multiple-scattering theory can be set up. Assume now that there are  $N$  plane scatterers placed at arbitrary positions. Assume them to be parallel, so that  $s$ - and  $p$ -polarized light do not mix in the scattering process. Consequently, an  $N$ -plane T-matrix can be determined for  $s$ - and  $p$ -polarized light separately. The  $N$ -plane T-matrix for  $s$ -waves is equal to the  $N$ -plane T-matrix for scalar waves, which was derived in the previous chapter. Here, we can concentrate on the  $N$ -plane T-matrix of  $p$ -polarized light. We repeat the formal sum (3.16) for a T matrix of  $N$  scatterers

$$\mathbf{T}^{(N)} = \sum_{\alpha} \mathbf{T}_{\alpha} + \sum_{\beta \neq \alpha} \sum_{\alpha} \mathbf{T}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\alpha} + \sum_{\gamma \neq \beta} \sum_{\beta \neq \alpha} \sum_{\alpha} \mathbf{T}_{\gamma} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\beta} \cdot \mathbf{G}_0 \cdot \mathbf{T}_{\alpha} + \dots \quad (3.43)$$

In Eq. (3.43) the Green functions are always sandwiched between T-matrices of planes at different positions. The values of  $\mathbf{G}_0(\mathbf{k}_{\parallel}, z_{\beta}, z_{\alpha}, \omega)$  with  $z_{\beta} \neq z_{\alpha}$  are finite. Therefore, once the single-plane  $T$  matrix is defined with the help of a regularization, it is not necessary to perform another regularization in order to find the  $N$ -plane T-matrix. One could argue that for consistency all Green functions in (3.43) should be replaced by the regularized ones, for without the regularization the T matrices would not even be defined. In practice, however, the differences between  $\mathbf{G}_0$  and  $\tilde{\mathbf{G}}_0$  can be neglected, because the planes are assumed to be at optical distances apart such that  $\Lambda|z_{\alpha} - z_{\beta}| \gg 1$  even for the planes  $\alpha$  and  $\beta$  closest to each other.

With the explicit form for the Green tensor and the T-matrices known, the series (3.43) can be summed exactly, each component separately and analogously to the scalar case in the previous chapter. This gives the central result of this section, the  $N$ -plane T-matrix for scattering by vector waves:

$$\mathbf{T}^{(N)}(\omega) = \frac{1}{(2\pi)^2} \int d^2\mathbf{k}_{\parallel} \sum_{\alpha, \beta=1}^N |\mathbf{k}_{\parallel}, z_{\alpha}\rangle \langle \mathbf{k}_{\parallel}, z_{\beta}| \mathbf{T}_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) \quad (3.44)$$

The only two nonzero spatial components of the  $3 \times 3$   $T$  matrices  $\mathbf{T}_{\alpha\beta}^{(N)}$  are

$$\begin{aligned} T_{\alpha\beta}^{ss, (N)}(\mathbf{k}_{\parallel}, \omega) &= T^{ss}(\mathbf{k}_{\parallel}, \omega) [\mathbb{1} - \mathcal{D}_s(\mathbf{k}_{\parallel}, \omega) T^{ss}(\mathbf{k}_{\parallel}, \omega)]_{\alpha\beta}^{-1} \\ T_{\alpha\beta}^{vv, (N)}(\mathbf{k}_{\parallel}, \omega) &= T^{vv}(\mathbf{k}_{\parallel}, \omega) [\mathbb{1} - \mathcal{D}_v(\mathbf{k}_{\parallel}, \omega) T^{vv}(\mathbf{k}_{\parallel}, \omega)]_{\alpha\beta}^{-1} \end{aligned} \quad (3.45)$$

The  $N^2$  matrix elements  $(\mathcal{D}_j)_{\alpha\beta}$  are defined as  $(1 - \delta_{\alpha\beta})G_0^{jj}(\mathbf{k}_{\parallel}, z_{\alpha}, z_{\beta}, \omega)$ , for  $j = s, v$ . The calculation  $\mathbf{T}^{(N)}$  boils down to the inversion of an  $N \times N$  matrix for the two transverse polarization directions separately. The  $ss$ -component is identical to the T-matrix  $T^{(N)}$  for scalar waves.

The trick of the analytical inversion of  $T^{ss,(N)}$  that was presented in appendix A and used in the previous chapter can be accomplished for  $T^{vv,(N)}$  as well:  $\nu = \nu_s \equiv -2ik_z/T^{ss}(\mathbf{k}_{\parallel}, \omega)$  should be replaced by  $\nu_p \equiv -2i(\omega/c)^2/[k_z T^{vv}(\mathbf{k}_{\parallel}, \omega)]$ . The derivation will not be repeated here, but we will give some results. In the first place, the Bloch wave vector  $K_p$  for  $p$ -polarized light reflected by planes separated by a distance  $a$  is given by  $\arccos(C_p)/a$ , with

$$C_p = \cos(k_z a) + \left( \frac{ik_z [k_z |T^{vv}|^2 + 2(\omega/c)^2 \text{Im} T^{vv}] + 2k_z (\omega/c)^2 \text{Re} T^{vv}}{k_z^2 (\text{Re} T^{vv})^2 + [2(\omega/c) + k_z \text{Im} T^{vv}]^2} \right) \sin(k_z a). \quad (3.46)$$

In general,  $C_p$  is a complex constant. However, if there is no light absorption in the planes so that the optical theorem (3.41) holds, then the imaginary part of  $C_p$  becomes identically zero. (For  $s$ -polarization we saw the same thing happening: without absorption,  $C_s$  is real.) For  $p$ -polarized light with wave vector and frequency such that  $|C_p| > 1$ , the Bloch wave vector is complex. The system of planes has a stop band for this light, meaning that the light will be 100% reflected when falling on a semi-infinite system of planes. If the optical theorem indeed holds, then the single-plane T-matrix is of the form (3.42) and the expression (3.46) can be simplified to give

$$C_p = \cos(k_z a) - \frac{k_z c^2 F_p(\mathbf{k}_{\parallel}, \omega)}{2\omega^2} \sin(k_z a). \quad (3.47)$$

### 3.2.7 A model for the optical potential

The most general T-matrix for  $p$ -waves (3.42) features an as yet unspecified optical potential that should be real when energy is conserved but for the rest it can be an arbitrary function of the in-plane wave vector and frequency. Until now we assumed that the optical potential  $F(\mathbf{k}_{\parallel}, \omega)$  was only frequency-dependent and we called it  $-V(\omega)$ , thereby neglecting spatial dispersion and anisotropy that would show up as a  $k_{\parallel}$ - and  $\hat{\mathbf{k}}_{\parallel}$ -dependence, respectively. Both were neglected as early as in the wave equation (3.5).

In chapter 2 plane scatterers were introduced as a simplified model for dielectric slabs of finite thickness  $d$  and nondispersive dielectric function  $\varepsilon(\omega) = \varepsilon$ . Recall that the optical potential for the plane scatterer in this model is obtained via the limiting process of making the dielectric slab thinner and increasing the polarizability  $\varepsilon - 1$  while keeping their product constant and equal to an “effective thickness”. Now for vector waves we use the same procedure and for both  $s$ - and  $p$ -polarized light we find the optical potential  $F(\mathbf{k}_{\parallel}, \omega) = -V(\omega) = D_{\text{eff}}(\omega/c)^2$ , with the effective thickness  $D_{\text{eff}}$  equal to  $d[\varepsilon(\omega) - 1]$ .

One can hope that transmission and reflection properties of a plane scatterer and a finite dielectric slab do not differ much. In this respect,  $p$ -polarized light is different from  $s$ -polarized light. When light in a medium with refractive index  $n_1$  meets an interface with a second medium with refractive index  $n_2 > n_1$ , then there is a typical angle of incidence,

called the Brewster angle, for which all  $p$ -polarized light is reflected. (For  $s$ -polarized light there is no such angle.) The Brewster angle  $\theta_B$  depends on the refractive-index contrast in the following way:

$$\theta_B = \tan^{-1}(n_2/n_1). \quad (3.48)$$

In the limiting procedure for going from a finite slab-in-air to an infinitely thin plane-in-air, the dielectric contrast  $\sqrt{\varepsilon}/1$  is going to infinity and consequently the Brewster angle becomes  $90^\circ$  in that limit, so that effectively the Brewster effect becomes invisible. Therefore, in our limiting procedure, a plane scatterer will not have a Brewster angle at the same angle as the finite dielectric slab that one tries to model with it.

### 3.3 Optical modes and omnidirectional mirrors

#### 3.3.1 Propagating modes

The optical modes are the harmonic solutions of the wave equation (3.5). These modes are also the solutions of the Lippmann-Schwinger equation, which is solved once the T-matrix of the total scattering system is found:

$$\mathbf{E}_{\mathbf{k}\sigma}(\mathbf{k}_{\parallel}, z, \omega) = E_0 \sigma_{\mathbf{k}} e^{ik_z z} + \sum_{\alpha, \beta} \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \mathbf{T}_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) \cdot \sigma_{\mathbf{k}} E_0 e^{ik_z z_{\beta}}. \quad (3.49)$$

The modes that correspond to an incoming plane wave ( $E_0 \neq 0$ ) are the propagating modes (or: radiative modes) and they are labelled by the incoming wave vector  $\mathbf{k}$  and polarization  $\sigma_{\mathbf{k}}$ . The  $s$ -polarized modes [with  $\sigma_{\mathbf{k}} = (1, 0, 0)$ ] are identical to the modes for scalar waves derived in the previous chapter. The  $p$ -polarized modes [ $\sigma_{\mathbf{k}} = (0, k_z/k, -k_{\parallel}/k)$ ] are the new ones.

Mode functions are complex functions of position and mode profiles are their absolute values squared. Mode profiles for  $s$ -polarized light are identical to the mode profiles of scalar waves that were discussed in chapter 2. We choose the light as incoming from the left. If in particular we choose light of frequency such that  $a/\lambda = 0.5$ , then the mode profiles of  $s$ -polarized light will correspond to the scalar mode profile of the same frequency in figure 2.2. For perpendicularly incident light, there is no difference between  $s$ - and  $p$ -polarization. In figure 3.1 the mode profiles for  $s$ - and  $p$ -polarized light inside a ten-plane crystal are compared both for an incoming angle of  $30^\circ$  and for  $60^\circ$ . In figure 3.1(a) we see that at an angle of  $30^\circ$  the mode profiles corresponding to both polarizations do not differ much yet. Both modes decay rapidly inside the crystal structure and are reflected (almost) completely. Only for the  $s$ -wave the polarization directions of the incoming and the reflected wave are equal, so that the amplitude of its mode profile at the left side of the crystal is four times the amplitude of the incoming electric field. The Bloch wave vectors are imaginary for both polarizations.

The situation is different at an incoming angle of  $60^\circ$  as shown in figure 3.1(b): there the mode profile of the  $s$ -polarized light again rapidly decays inside the crystal (and the corresponding Bloch wave vector will again be imaginary), whereas the  $p$ -polarized light

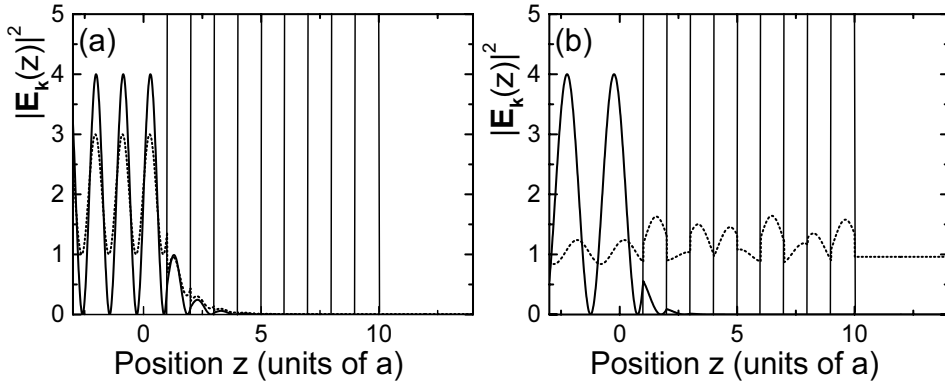


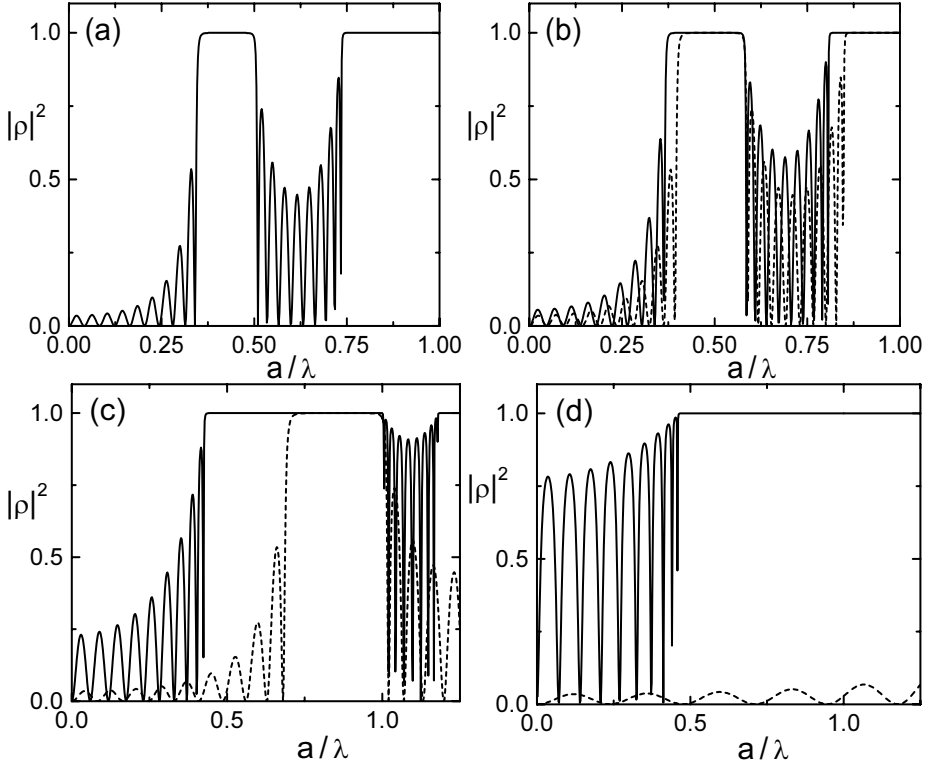
**Figure 3.1:**

Squares of absolute values of mode functions for  $s$ -polarized (solid lines) and  $p$ -polarized light (dashed lines), as a function of position. The light is scattered by a crystal of ten planes with  $D_{\text{eff}} = 0.46a$ , separated by a distance  $a$ . Both modes correspond to light incoming from the left with  $a/\lambda = 0.5$ . Figure (a):  $\theta_{\text{in}} = 30^\circ$ ; figure (b):  $\theta_{\text{in}} = 60^\circ$ .

can propagate inside the crystal (so the Bloch wave vector is real) and the light is transmitted almost completely. For this frequency and incoming angle, the crystal is a good polarization filter.

Note in figures 3.1(a,b) that the mode profiles of the  $s$ -polarized waves are continuous whereas  $p$ -polarized waves form more irregular patterns and are discontinuous at the positions of the planes. This reflects the boundary conditions that are automatically satisfied by the modes that we find by multiple-scattering theory: the tangential components of the electric fields must be continuous and the normal components show a jump at a dielectric interface. Now the electric field of  $s$ -polarized light only has a tangential component and  $p$ -polarized light consists of both a tangential and a normal component. This explains the differences in the mode profiles for  $s$ - and  $p$ -waves.

One can also compare reflection by the ten-plane Bragg mirror as a function of frequency for the two polarization directions. This is plotted in figure 3.2. For light incident perpendicularly to the planes, both transverse polarization vectors are equivalent and accordingly in figure 3.2(a),  $s$ - and  $p$ -polarized light show identical reflection characteristics as a function of frequency. This reflection plot is identical to the one for scalar waves in the previous chapter. Differences between the two polarizations do appear for non-normal incidence. In figure 3.2(b) the incident angle is  $30^\circ$  and in figures (c)-(d) the incident angle is increased further. The red edges of the stop bands for  $s$ -polarized light move to higher frequencies and the widths of the stop bands become larger. For  $p$ -polarized light on the other hand, the red edges of the stop bands also shift to the blue, and faster so for larger incident angles. In contrast, the stop bands for  $p$ -light become narrower while moving to higher frequencies. In figure (d) for an incident angle of  $80^\circ$ , the first stop band for  $p$ -polarized light has even moved out of the picture, while the  $s$ -stop band has become very broad. It is harder for the ten-plane crystal to stop  $p$ -polarized light than to stop  $s$ -polarized

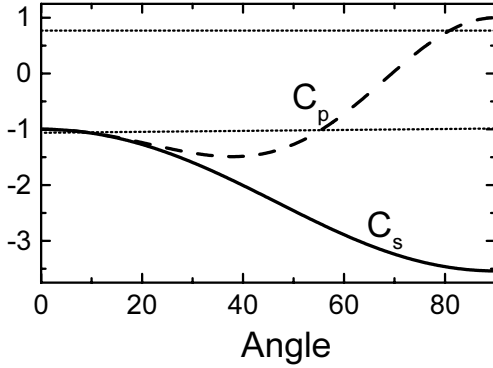
**Figure 3.2:**

Reflection off a ten-plane crystal, as a function of  $a/\lambda$ , for  $s$ -polarized light (solid lines) and  $p$ -polarized light (dashed lines). Incoming angles are  $0^\circ$ ,  $30^\circ$ ,  $60^\circ$  and  $80^\circ$ . The planes have effective thickness  $D_{\text{eff}} = 0.46a$  and they are separated by a distance  $a$ .

light.

In the previous chapter we showed that a ten-plane crystal can act as an omnidirectional mirror for scalar waves. But is this crystal also an omnidirectional mirror for vector waves? For  $s$ -polarized light we know that there are frequency intervals in which light coming from all directions is reflected, because the  $s$ -waves map on the scalar waves. So here we need to analyze whether there are also such frequency intervals for  $p$ -waves and if so, whether any  $s$ - and  $p$ -frequency intervals have an overlap. Only in the latter case can we speak of an omnidirectional mirror for light.

It is the Bloch wave vector that distinguishes between light that can propagate inside a crystal and light that feels a stop band: a real Bloch wave vector corresponds to propagating light and a complex Bloch wave vector to light that is reflected off the crystal. In our formalism, the Bloch wave vectors are the arc cosines of the constants  $C_s$  and  $C_p$ . Now for light of a frequency corresponding to  $a/\lambda = 0.5$  and planes with  $D_{\text{eff}} = 0.46a$  we know that  $s$ -waves are reflected omnidirectionally. In figure 3.3 we plot both constants for

**Figure 3.3:**

Constants  $C_s$  and  $C_p$  as a function of angle of the incoming light, for the parameters  $a/\lambda = 0.5$  and  $D_{\text{eff}} = 0.46a$ . Regions where  $-1 \leq C \leq 1$  correspond to propagating waves inside the crystal.

this frequency, as a function of angle of the incident light. Unlike for  $s$ -waves, for  $p$ -waves there are incident angles larger than the critical angle  $\theta_c \gtrsim 55^\circ$  for which the values of  $C_p$  are between -1 and 1. Light incident with these large angles can propagate inside the crystal and therefore the crystal is not an omnidirectional mirror for this frequency. Actually, this information could already be read off from the mode profile in figure 3.1(b). However, the conclusion does not only hold for the specific parameter values that we chose: if we choose  $D_{\text{eff}}$  larger then  $\theta_c$  moves up to larger angles, but it can be shown by expanding Eq. (3.47) around  $\theta_{\text{in}} = 90^\circ$  that for every finite  $D_{\text{eff}}$  and  $a/\lambda$  there always is a finite interval of angles corresponding to propagating  $p$ -polarized light. So strictly speaking, omnidirectional mirrors for vector waves can not be made with equidistant identical plane scatterers, but it is possible to block incident light from almost all directions.

### 3.3.2 Guided modes

We have found the propagating modes of vector waves near plane scatterers but these do not necessarily form the complete set of modes. As pointed out in section 3.1, a scattering problem can also have bound modes that do not correspond to incoming light. By solving the Lippmann-Schwinger equation without an incoming field, modes of scalar waves bound or guided by one or more plane scatterers were found in section 2.4.2. Later on, in section 2.5, it was shown that with each guided mode a nonzero local density of states is associated. One can also turn it around and try and find guided modes by looking for values of  $k_{\parallel} > \omega/c$  corresponding to a nonzero density of states. It is the latter approach that we follow here for vector waves, because it directly allows us to use the effective description (3.36) for the T-matrix.

For vector waves, the local density of states is a tensor proportional to the imaginary part of the Green tensor  $\mathbf{G}(\mathbf{r}, \mathbf{r}, \omega)$ . In planar geometries the latter can best be found as an integral over the Green tensor in the plane representation:

$$\mathbf{G}(\mathbf{r}, \mathbf{r}, \omega) = \frac{1}{(2\pi)^2} \int d^2\mathbf{k}_{\parallel} \mathbf{G}(\mathbf{k}_{\parallel}, z, z, \omega). \quad (3.50)$$

Clearly, the local density of states can only be nonzero if the imaginary part of the integrand in (3.50) is nonzero. A guided mode manifests itself when this integrand has a

nonzero imaginary part for a certain  $k_{\parallel} > \omega/c$ . For the crystals of plane scatterers the Green tensor directly follows from the Dyson-Schwinger equation:

$$\mathbf{G}(\mathbf{k}_{\parallel}, z, z', \omega) = \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega) + \sum_{\alpha, \beta=1}^N \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \mathbf{T}_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) \cdot \mathbf{G}_0(\mathbf{k}_{\parallel}, z_{\beta}, z', \omega). \quad (3.51)$$

All the components of  $\mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega)$  become real quantities for  $k_{\parallel} > \omega/c$ . This means that there is no density of states corresponding to guided modes in free space, and this comes as no surprise. Therefore, equation (3.51) tells us that the imaginary part of the Green tensor  $\mathbf{G}$  can only be nonzero when at least one of the two spatial components of the  $N$ -plane T matrix  $\mathbf{T}^{(N)}$  has a pole. These components  $T_{\alpha\beta}^{ss, (N)}(\mathbf{k}_{\parallel}, \omega)$  and  $T_{\alpha\beta}^{vv, (N)}(\mathbf{k}_{\parallel}, \omega)$  are given in equation (3.45).

First look for the poles of the component  $T_{\alpha\beta}^{ss, (N)}(\mathbf{k}_{\parallel}, \omega)$ . This is easy, because this component is identical to the  $N$ -plane T matrix  $T^{(N)}$  for scalar waves, as presented in the previous chapter. So we find that there are at most  $N$  guided modes corresponding to  $s$ -polarized light in a crystal of  $N$  planes. The dispersion relations of these guided modes are given in figure 2.4.

Are there also guided modes corresponding to  $p$ -polarized light in a crystal of  $N$  planes? This question will be addressed in some detail now. We need to find the poles of the component  $T_{\alpha\beta}^{vv, (N)}(\mathbf{k}_{\parallel}, \omega)$ . They occur when the determinant  $\det[(\mathbf{T}^{vv, (N)})^{-1}]$  is equal to zero. An expression for this determinant can be found if one replaces  $\nu_s$  by  $\nu_p$  (as defined in section 3.2.6) in the expression A.14 for the corresponding determinant for  $s$ -polarized light. The result is that for  $p$ -polarized light a guided mode exists when the following equation is satisfied:

$$2(\omega/c)^2 \sin[(N+1)K_p a] + [\kappa F - 2(\omega/c)^2] e^{-\kappa a} \sin(NK_p a) = 0, \quad (3.52)$$

where  $\kappa$  is defined as  $-i\sqrt{(\omega/c)^2 - k_{\parallel}^2}$ . The Bloch wave vector  $K_p$  is still defined as  $a^{-1}$  times the arc cosine of the constant  $C_p$  (Eq. (3.47)), but in terms of  $\kappa$  it reads

$$C_p = \cosh(\kappa a) + \frac{\kappa c^2 F}{2\omega^2} \sinh(\kappa a). \quad (3.53)$$

Equation (3.52) should lead to the dispersion relations  $\omega(\kappa)$  for the guided modes, if they exist. When increasing the frequency, new guided modes appear that at first are only just captured by the structure so that  $\kappa = 0^+$ . As we did for  $s$ -polarized guided modes, we look for the guided modes in this small- $\kappa$  limit. Note that for  $\kappa$  very small and positive, the constant  $C_p$  can be written as  $1 + (\chi\kappa)^2/2$ , up to second order in  $\kappa$ , with  $\chi$  defined as  $a\sqrt{1 + Fc^2/(2a\omega^2)}$ . We can estimate the Bloch wave vector for small  $\kappa$  as follows:

$$C_p = 1 + (\chi\kappa)^2/2 + o(\kappa^3) = \cosh(\chi\kappa) + o(\kappa^3) \Rightarrow K_p = i\chi\kappa/a \quad (3.54)$$

It follows that for small and positive  $\kappa$ , solutions of equation (3.52) will only exist when  $\sinh[(N+1)\chi\kappa]$  equals  $\sinh(N\chi\kappa)$ , or equivalently  $\chi = 0$ . If for the optical potential we take  $F = D_{\text{eff}}(\omega/c)^2$ , then we find that there are guided modes for  $p$ -polarized light

whenever  $1 + D_{\text{eff}}/(2a) = 0$ . So in a crystal of  $N$  planes with a positive effective thickness, there are no guided modes corresponding to  $p$ -polarized light. This is in agreement with the result in [65] for infinite crystals.

In conclusion, all guided modes in the finite crystal of plane scatterers correspond to  $s$ -polarized light and their properties have already been studied in the previous chapter. For dielectric slabs of finite thickness it is known that  $s$ -waves can be guided more easily than  $p$ -waves [60]. The non-existence of guided  $p$ -waves for plane scatterers is a manifestation of this difference between the two polarization directions.

## 3.4 Spontaneous emission

### 3.4.1 General theory applied to planes

The spontaneous emission rate  $\Gamma$  of an atom embedded in an inhomogeneous dielectric is given by [11]

$$\Gamma(\mu, \mathbf{R}, \Omega) = \pi \sum_l \frac{\omega_l}{2\hbar\varepsilon_0} |\mu \cdot \mathbf{E}_l(\mathbf{R})|^2 \delta(\omega_l - \Omega). \quad (3.55)$$

This can be found by applying Fermi's golden rule. The spontaneous-emission rate (3.55) depends on the atomic transition frequency  $\Omega$ , on the  $z$ -coordinate of its position  $\mathbf{R} = (x, y, z)$  and on the magnitude and orientation of its dipole moment  $\mu$ . The  $\mathbf{E}_l$  are the normal mode solutions of the wave equation (3.5) for the electric field. The spontaneous-emission rate can alternatively be expressed in terms of the Green function of the medium. A derivation will be presented in section 5.3.1, but the rate to be found in Eq. (5.36) will already be used now:

$$\Gamma(\mu, \mathbf{R}, \Omega) = -\frac{2\Omega^2}{\hbar\varepsilon_0 c^2} \text{Im} [\mu \cdot \mathbf{G}(\mathbf{R}, \mathbf{R}, \Omega) \cdot \mu]. \quad (3.56)$$

Here,  $\mathbf{G}$  is the dyadic Green function of the electric-field wave equation (3.5). For systems consisting of dielectric layers (not necessarily plane scatterers), it is easiest to first calculate the Green function in the plane representation  $\mathbf{G}(\mathbf{k}_{\parallel}, z, z, \Omega)$ . We only need to Fourier transform this Green function to real space as in Eq. (3.50) in order to find the Green function of Eq. (3.56) that determines the spontaneous-emission rates. A slight complication is that until now we chose a representation for  $\mathbf{G}(\mathbf{k}_{\parallel}, z, z, \Omega)$  which is co-rotating with the incoming wave vector  $\mathbf{k}_{\parallel}$  that we now want to integrate over. We need a fixed basis  $\{\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}\}$  and we can choose it such that the atomic dipole becomes  $(\mu_x, 0, \mu_z)$  in the new representation. We perform the two-dimensional integral  $\int d^2\mathbf{k}_{\parallel}$  by writing it in polar coordinates  $\int_0^\infty dk_{\parallel} k_{\parallel} \int_0^{2\pi} d\hat{\mathbf{k}}_{\parallel}$ . By doing the angular integral first, only diagonal elements of the dyadic Green function survive. The total spontaneous emission-rate is the sum of two contributions, the perpendicular and the parallel decay rate, which are given

by, respectively,

$$\Gamma_z(z, \Omega) = -\frac{3c\Gamma_0\mu_z^2}{\Omega\mu^2} \text{Im} \int_0^\infty dk_{\parallel} k_{\parallel} G^{zz}(k_{\parallel}, z, z, \Omega) \quad (3.57)$$

$$\Gamma_x(z, \Omega) = -\frac{3c\Gamma_0\mu_x^2}{\Omega\mu^2} \text{Im} \int_0^\infty dk_{\parallel} k_{\parallel} \frac{G^{ss}(k_{\parallel}, z, z, \Omega) + G^{vv}(k_{\parallel}, z, z, \Omega)}{2}. \quad (3.58)$$

The parallel decay-rate has a contribution both from  $s$ - and  $p$ -polarized light ( $G^{ss}$  and  $G^{vv}$ , respectively) whereas the perpendicular decay-rate only has a  $p$ -polarized decay channel (through  $G^{zz}$ ). When the dipole is oriented parallel to the plane, the perpendicular decay-rate is zero, and vice versa. The spontaneous-emission rates in Eq. (3.57) and (3.58) are integrals over all possible lengths of the in-plane wave vector. Both rates can be subdivided into a propagating (or radiative) rate corresponding to the integration of  $k_{\parallel}$  from 0 to  $\omega/c$ , and a guided rate which is the integral from  $\omega/c$  to infinity.

More precisely, we can subdivide the parallel decay-rate into three parts: an  $s$ -polarized radiative rate ( $sr$ ), a  $p$ -polarized radiative rate ( $pr$ ) and an  $s$ -polarized guided rate ( $sg$ ). There are no guided modes for  $p$ -polarized light and as a consequence the Green tensor  $G^{zz}$  has no imaginary part (zero density of states) for  $k_{\parallel} > \omega/c$ . The perpendicular decay-rate  $\Gamma_z$  is therefore purely radiative:

$$\Gamma_x^{sr}(z, \omega) = -\frac{3c\Gamma_0}{2\Omega} \left(\frac{\mu_x}{\mu}\right)^2 \text{Im} \int_0^{\omega/c} dk_{\parallel} k_{\parallel} G^{ss}(k_{\parallel}, z, z, \Omega), \quad (3.59a)$$

$$\Gamma_x^{pr}(z, \Omega) = -\frac{3c\Gamma_0}{2\Omega} \left(\frac{\mu_x}{\mu}\right)^2 \text{Im} \int_0^{\omega/c} dk_{\parallel} k_{\parallel} G^{vv}(k_{\parallel}, z, z, \Omega), \quad (3.59b)$$

$$\Gamma_x^{sg}(z, \Omega) = -\frac{3c\Gamma_0}{2\Omega} \left(\frac{\mu_x}{\mu}\right)^2 \text{Im} \int_{\omega/c}^\infty dk_{\parallel} k_{\parallel} G^{ss}(k_{\parallel}, z, z, \Omega), \quad (3.59c)$$

$$\Gamma_z(z, \Omega) = -\frac{3c\Gamma_0}{\Omega} \left(\frac{\mu_z}{\mu}\right)^2 \text{Im} \int_0^{\omega/c} dk_{\parallel} k_{\parallel} G^{zz}(k_{\parallel}, z, z, \Omega). \quad (3.59d)$$

With all the spontaneous-emission rates spelled out now, let us first study spontaneous-emission rates near a single plane.

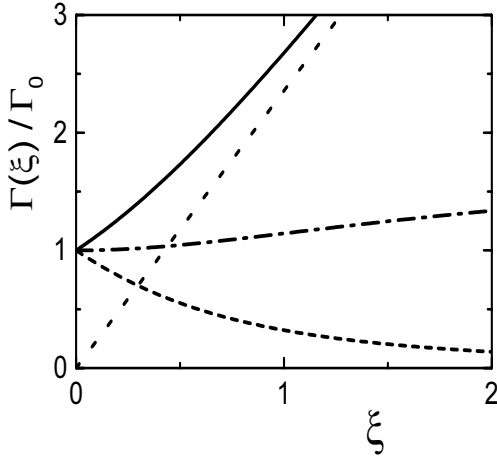
### 3.4.2 Spontaneous emission near one plane scatterer

The Green function in the plane representation can be calculated directly using the single-plane T-matrix  $\mathbf{T}$  of section 3.2.6 in the Dyson-Schwinger equation

$$\mathbf{G}(\mathbf{k}_{\parallel}, z, z', \omega) = \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega) + \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \mathbf{T}(\mathbf{k}_{\parallel}, \omega) \cdot \mathbf{G}_0(\mathbf{k}_{\parallel}, z_{\alpha}, z', \omega). \quad (3.60)$$

This Green function can be used in the expressions of the previous section 3.4.1 to obtain spontaneous-emission rates near a single plane.

What happens to the spontaneous-emission rates very close to the plane? (By “close” it is meant: at distances small compared to the wavelength of light, but still large compared to atomic distances where local-field effects come into play that we do not consider

**Figure 3.4:**

Spontaneous-emission rates of dipoles with zero distance to the single plane scatterer, as a function of the dimensionless parameter  $\xi = \pi D_{\text{eff}}/\lambda$ . Solid line: the rate  $\Gamma_x$ , which is the sum of a rate into radiative (dashed line) and into guided modes (dotted line). The rate  $\Gamma_z$  is the dash-dotted line.

here.) In the limit that the atomic position  $z$  becomes equal to the plane position  $z_\alpha$ , the spontaneous-emission rates for the two orientations can be calculated analytically. Let  $\Gamma_0$  be the spontaneous-emission rate in vacuum, equal to  $\mu^2 \Omega^3 / (3\pi \hbar \varepsilon_0 c^3)$ . For a dipole perpendicular to the planes we find

$$\Gamma_z(z_\alpha, \Omega) = \Gamma_0 \left( 2 + \frac{3}{2\xi^2} \right) - \frac{3}{2}\Gamma_0 \left( 1 + \frac{1}{\xi^2} \right) \frac{\arctan(\xi)}{\xi}, \quad (3.61)$$

where the dimensionless parameter  $\xi$  is defined as  $\pi D_{\text{eff}}/\lambda$ . The three contributions to the parallel decay-rate at the position of the plane can also be expressed in terms of the parameter  $\xi$  alone:

$$\Gamma_x^{sr}(z_\alpha, \Omega) = \frac{3}{4}\Gamma_0 [1 - \xi \arctan(1/\xi)], \quad (3.62a)$$

$$\Gamma_x^{pr}(z_\alpha, \Omega) = \frac{3}{4\xi^2}\Gamma_0 \left[ 1 - \frac{\arctan(\xi)}{\xi} \right], \quad (3.62b)$$

$$\Gamma_x^{sg}(z_\alpha, \Omega) = \frac{3\pi\xi}{4}\Gamma_0. \quad (3.62c)$$

In figure 3.4 the relative rates are plotted as a function of  $\xi$ . The results can be checked in two limiting cases: the weakest test is the limit of invisible planes where  $D_{\text{eff}} = 0$  and therefore  $\xi = 0$ . Then both  $\Gamma_z$  and  $\Gamma_x$  approach the free-space value, as they should. The other test is the limit of a perfect mirror, when  $\xi = D_{\text{eff}} = \infty$ . This limit is not visible in the figure, but we find  $\Gamma_z/\Gamma_0 = 2$  and  $\Gamma_x/\Gamma_0 = 0$ . To be precise, we first have taken the limit of the perfect mirror and then come closer with the atom to the mirror. In doing so, the contribution to the spontaneous-emission rate of the guided waves goes to zero. Taking the limits in the reverse order would give the unphysical result that the contribution of the guided waves diverges when the positions of the atom and mirror coincide. The spontaneous-emission rates for atoms near perfect mirrors agree with the values in the literature [16, 78]. These results can be explained in terms of constructive or destructive interference of the light emitted by the atom and its perfect mirror image.

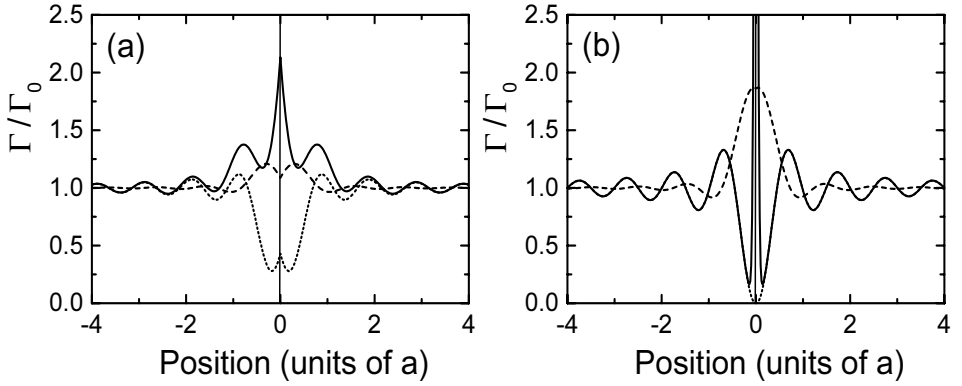


**Figure 3.5:**

Spontaneous-emission rates of dipoles near a single partially transmitting plane scatterer, relative to  $\Gamma_0$ . Solid lines correspond to total spontaneous-emission rates  $\Gamma_x$  for dipoles parallel to the plane, dotted lines are radiative contributions to  $\Gamma_x$ , and the dashed lines denote  $\Gamma_z$ . In (a), the effective thickness  $D_{\text{eff}}$  of the plane equals  $0.46a$  and in (b)  $D_{\text{eff}} = 10a$ .

In our model, effects on spontaneous emission become more pronounced when the parameter  $\xi$  is larger and this can be accomplished by choosing a higher frequency. However, this only makes sense physically in the frequency regime where we can neglect dispersion in the dielectric function  $\varepsilon(\omega)$  of the dielectric slabs that are being modelled. Otherwise, the dispersion shows up in the effective thickness  $D_{\text{eff}} = d[\varepsilon(\omega) - 1]$ , with the effect that there is a frequency for which the parameter  $\xi$  is maximal. Dielectrics do not make perfect mirrors for high frequencies. Theoretically, this point is important for a consistent description of quantum optics in dielectrics [79].

In figure 3.5(a), spontaneous-emission rates as a function of position are plotted for  $D_{\text{eff}} = 0.46a$ . The values for  $z = 0$  are finite in both cases (although not shown in figure 3.5) and indeed correspond to Eqs. (3.61) and (3.62a). For both dipole orientations, far away from the plane the rate approaches the free-space value. Close to the plane,  $\Gamma_z$  is increased; the  $\Gamma_x$  is also larger than  $\Gamma_0$ , but this decay-rate consists of a rate into propagating modes that is less than  $\Gamma_0$  and a guided-mode-rate. The contributions of radiative and guided  $s$ -waves for an atom with  $\mu = \mu_x$  are the same as for scalar waves with “scalar dipole moment”  $\mu$ , but since the total decay-rate  $\Gamma_0$  for vector waves is larger than for scalar waves, the relative contributions of  $s$ -waves to  $\Gamma/\Gamma_0$  are smaller for vector waves (by a factor  $3/4$ ). In figure 3.5(b), the same rates are plotted, but now for a much better reflecting plane with an effective thickness of  $D_{\text{eff}} = 10a$ . There the limiting values of spontaneous-emission rates near the plane are approached:  $\Gamma_z$  climbs up but does not reach the value of  $2\Gamma_0$  yet, whereas the propagating part of  $\Gamma_x$  is practically zero when  $D_{\text{eff}}$  equals  $10a$ . The partial emission-rate into the guided mode has a much larger amplitude near the plane but falls off much more rapidly away from the plane.

### 3.4.3 Spontaneous emission near a ten-plane scatterer

In our formalism we can study spontaneous-emission rates in the vicinity of an arbitrary number of planes. Here we choose to present results for  $N = 10$ , as we did for scalar waves. The spontaneous-emission rates calculated in this section are based on Eqs. (3.59a-3.59d). The following Dyson-Schwinger equation gives the relevant Green functions in terms of the T-matrix of section 3.2.6:

$$\mathbf{G}(\mathbf{k}_{\parallel}, z, z', \omega) = \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z', \omega) + \sum_{\alpha, \beta=1}^N \mathbf{G}_0(\mathbf{k}_{\parallel}, z, z_{\alpha}, \omega) \cdot \mathbf{T}_{\alpha\beta}^{(N)}(\mathbf{k}_{\parallel}, \omega) \cdot \mathbf{G}_0(\mathbf{k}_{\parallel}, z_{\beta}, z', \omega). \quad (3.63)$$

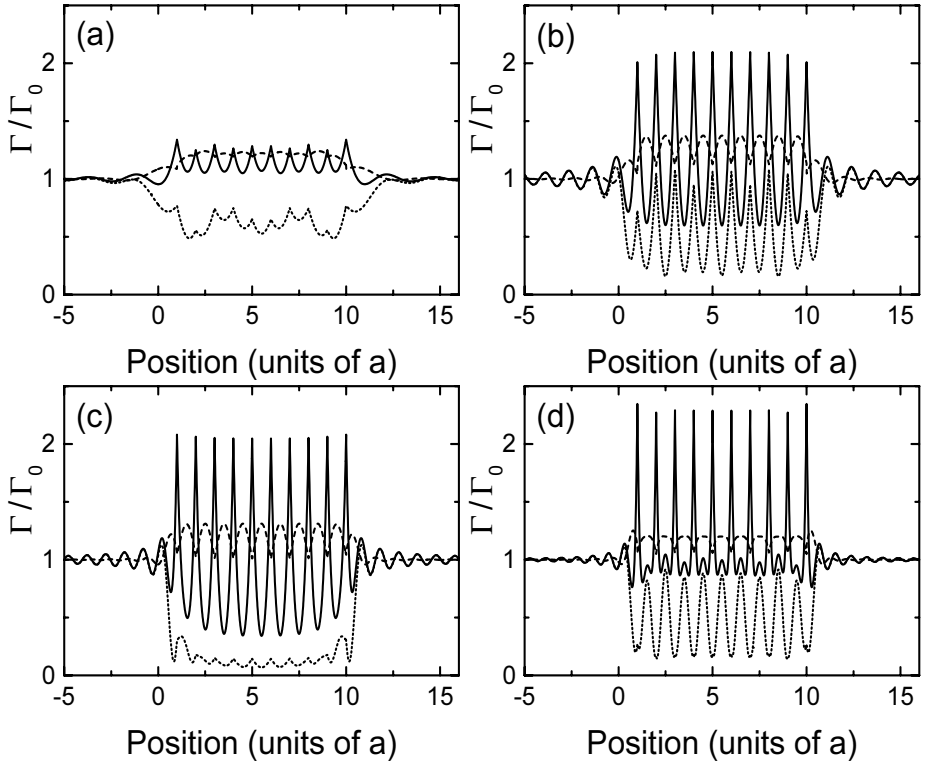
The first thing that one should like to do in a formalism for vector waves is to compare the total emission rates for dipoles parallel ( $\Gamma_x$ , solid lines) with dipoles perpendicular ( $\Gamma_z$ , dashed lines) to the planes. In figure 3.6(a-d) orientation-dependent spontaneous-emission rates are plotted for several frequencies. For clarity in the pictures, the positions of the planes at 1, 2, ..., 10 are not shown as vertical lines this time. The most striking difference is that  $\Gamma_x$  becomes very spiky near the planes because of the guided modes to which only parallel dipoles can couple in the vicinity of the planes. We can also see that  $\Gamma_z$ , which is purely radiative, on average increases due to the presence of the planes whereas the radiative part of  $\Gamma_x$  on average decreases near the planes. In the previous section we saw the same trend near a single plane where in the limit of a perfect mirror  $\Gamma_x/\Gamma_0 \rightarrow 0$  and  $\Gamma_z/\Gamma_0 \rightarrow 2$ . Figure 3.1 showed that the optical modes of  $p$ -polarized light have discontinuities at the plane positions. There are also discontinuities in the spontaneous-emission rates, but these discontinuities are too small to be visible in figure 3.6. They are small because the discontinuities per mode are averaged in the emission rate.

The dotted lines in figure 3.6 are the radiative parts of  $\Gamma_x$ . These are similar to the radiative rates for scalar waves, but not identical since in  $\Gamma_x$  not only  $s$ -polarized light but also  $p$ -polarized light contributes. In particular, far away from the planes, the emission rate of dipoles parallel to the planes consists of 75 percent  $s$ -polarized and 25 percent  $p$ -polarized light.

For scalar waves the ten-plane structure can act an omnidirectional mirror, whereas in section 3.3.1 it was found that it is not an omnidirectional mirror for vector waves. Correspondingly, the radiative LDOS for scalar waves at  $a/\lambda = 0.5$  dropped down to (almost) zero inside the ten-plane omni-directional mirror, whereas the emission rates in figure 3.6(c) show that the radiative LDOS for vector waves stays nonzero inside the crystal. In the inner unit cells, dipoles parallel to the planes emit predominantly guided light, but the small amount of light that leaves the structure is strongly  $p$ -polarized. This is the case around  $a/\lambda = 0.5$  only, where  $s$ -polarized light is omnidirectionally reflected; at higher and lower frequencies the emitted light can have both polarization directions.

## 3.5 Radiative line shifts

Not only spontaneous-decay rates change inside a dielectric medium, but there are also radiative shifts of the atomic transition frequencies. Both effects will be derived in detail

**Figure 3.6:**

Spontaneous-emission rates  $\Gamma_x$  (solid line) and  $\Gamma_z$  (dashed) near a ten-plane crystal. The dotted line is the radiative part of  $\Gamma_x$ . The figures (a)-(d) correspond to four frequencies: (a)  $a/\lambda = 0.2$ ; (b)  $a/\lambda = 0.4$ ; (c)  $a/\lambda = 0.5$ ; (d)  $a/\lambda = 0.6$ .

in section 5.3.1. As given in Eq. (5.38), the atomic transition frequency in a medium will be shifted with respect to the value  $\Omega$  in free space by an amount

$$\Delta(\mathbf{R}, \Omega) = \left( \frac{\mu^2 \Omega^2}{\hbar \epsilon_0 c^2} \right) \text{Re} \hat{\boldsymbol{\mu}} \cdot [\mathbf{G}(\mathbf{R}, \mathbf{R}, \Omega) - \mathbf{G}_0(\mathbf{R}, \mathbf{R}, \Omega)] \cdot \hat{\boldsymbol{\mu}}, \quad (3.64)$$

which depends both on position and frequency. In a T-matrix formalism, the Green function is the sum of the free-space Green function and the term  $\mathbf{G}_0 \cdot \mathbf{T} \cdot \mathbf{G}_0$ , see Eq. (3.63). Line shifts are therefore determined by the latter term only. Proceeding much the same way as for the decay rates in section 3.4.1, the line shift at position  $\mathbf{R} = (x, y, z)$  in a crystal of plane scatterers is found to be

$$\Delta(\mathbf{R}, \Omega) = \left( \frac{3c\Gamma_0}{2\Omega} \right) \text{Re} \left[ \frac{\mu_x^2}{2\mu^2} (I_s + I_v) + \frac{\mu_z^2}{\mu^2} I_z \right], \quad (3.65)$$

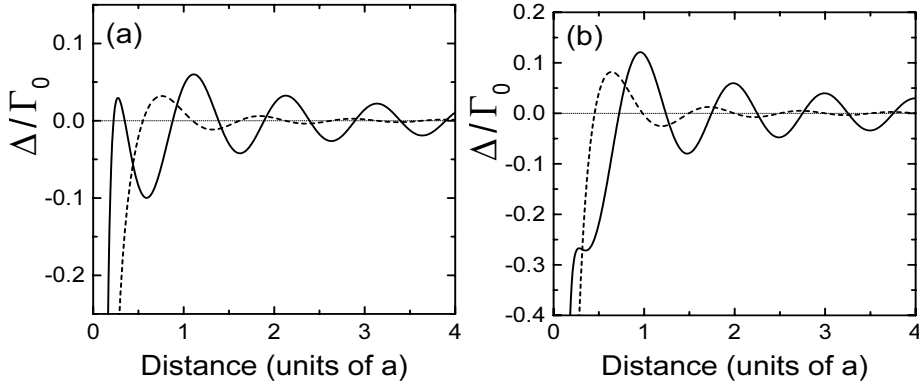


**Figure 3.7:**

Line shifts of the optical transition frequency relative to the free-space value  $\Omega$ . Shifts are given as a function of the distance of the atom to a single plane, both for atoms with dipole moments pointing parallel (solid lines) and perpendicular (dashed lines) to the plane. Shifts are scaled to the free-space decay rate  $\Gamma_0$ . In (a),  $D_{\text{eff}} = 0.46a$  and in (b)  $D_{\text{eff}} = 10a$ . In both figures,  $a/\lambda = 0.5$ .

where the integrals  $I_s$ ,  $I_v$  and  $I_z$  are given by

$$I_s = \sum_{\alpha\beta} \int_0^\infty dk_{\parallel} k_{\parallel} G_0^{ss}(k_{\parallel}, z, z_{\alpha}, \Omega) T_{\alpha\beta}^{ss}(k_{\parallel}, \Omega) G_0^{ss}(k_{\parallel}, z_{\beta}, z, \Omega), \quad (3.66)$$

$$I_v = \sum_{\alpha\beta} \int_0^\infty dk_{\parallel} k_{\parallel} G_0^{vv}(k_{\parallel}, z, z_{\alpha}, \Omega) T_{\alpha\beta}^{vv}(k_{\parallel}, \Omega) G_0^{vv}(k_{\parallel}, z_{\beta}, z, \Omega), \quad (3.67)$$

$$I_z = \sum_{\alpha\beta} \int_0^\infty dk_{\parallel} k_{\parallel} G_0^{zv}(k_{\parallel}, z, z_{\alpha}, \Omega) T_{\alpha\beta}^{vv}(k_{\parallel}, \Omega) G_0^{vz}(k_{\parallel}, z_{\beta}, z, \Omega). \quad (3.68)$$

In these integrals, the magnitude  $k_{\parallel}$  of the wave vector parallel to the planes goes from zero to infinity. The matrix elements  $T_{\alpha\beta}^s(k_{\parallel}, \omega)$  in the integral  $I_s$  have poles on the real- $k_{\parallel}$  axis corresponding to the guided modes. The real part of  $I_s$  can be evaluated by taking the Cauchy principal-value integral at these poles. Although there are no guided modes for  $p$ -polarized light, there is a nonzero contribution to the integral  $I_z$  from in-plane wave vectors larger than  $\Omega/c$ . There was no such contribution to the spontaneous-emission rate  $\Gamma_z$  in Eq. (3.59d). The difference stems from the fact that the decay rate is an on-shell quantity, whereas line shifts  $\Delta(\Omega)$  have off-shell contributions. In other words, unlike the rate  $\Gamma(\Omega)$ , the shift  $\Delta(\Omega)$  also depends on optical modes with eigenfrequencies different from  $\Omega$ . Actually, it is a weighted sum over all optical modes.

In figure 3.7(a), line shifts are presented as a function of distance to a single plane, for dipoles pointing parallel and perpendicular to the plane. Away from the plane, the shifts show damped oscillatory behavior. The magnitudes are very small, typically ten percent or less of the free-space decay rate. Close to the planes, the off-shell contributions of large wave vectors become important. For both dipole orientations, the line shifts even

diverge when approaching the plane. At distances on the order of  $0.1\lambda$  and smaller, the perpendicular dipoles feel larger frequency shifts than the parallel ones. On the other hand, far away from the plane the shifts are larger for parallel dipoles. Figure 3.7(b) shows shifts near a plane with much larger effective thickness. The amplitudes of the shifts are increased as compared to figure 3.7(a), but the overall picture has not changed.

In order to avoid the infinite line shifts at the position of the plane, the cutoff parameter  $\Lambda$  could have been kept finite. Then the discontinuities in modes and in Green functions would have become smooth. By keeping  $\Lambda \gg \Omega/c$  finite, only the line shifts at distances of typically  $\Lambda^{-1}$  or less are modified and become finite. The emission rates presented in this chapter already were finite and they will not be affected either at distances greater than  $\Lambda^{-1}$  when a finite cutoff is chosen to keep line shifts finite.

## 3.6 Conclusions and outlook

In this chapter a multiple-scattering theory was set up for the scattering of vector waves by parallel planes. The Green function had to be regularized and this was done by introducing a high-momentum cutoff. In the end, the regularization parameter was sent to infinity. An effective scattering theory for plane scatterers emerged with a nonzero T-matrix that no longer depends on the cutoff. This is in contrast with point scatterers where two cutoffs must be kept finite in order to define a nonzero  $T$  matrix [69]. The only reason to keep a finite cutoff for plane scatterers would be to avoid diverging line shifts at the positions of the planes.

The planar symmetry of the crystal of parallel planes enabled us to separate  $s$ - and  $p$ -polarized modes. The scattering by a nonabsorbing plane scatterer satisfies a separate optical theorem for each polarization. The radiative and guided modes of  $s$ -polarized light could be mapped onto modes for scalar waves. The  $s$ -polarized light has continuous mode functions, whereas the mode function of the  $p$ -polarized light is discontinuous at the positions of the planes. As a consequence, the  $p$ -polarization contributions to the spontaneous emission rate show discontinuities as well. Spontaneous emission rates were calculated, both for dipoles parallel and perpendicular to the planes.

One could wish to create a crystal such that 100% of the spontaneously emitted light is guided light. In our model of identical equidistant plane scatterers this will not happen. If there was only  $s$ -polarized light, then it would be possible, as it was for scalar waves [figure 2.7(a)]. Deep inside an omnidirectional mirror all spontaneously emitted light is guided light. For vector waves, however, equidistant plane scatterers can not be an omnidirectional mirror.

Omnidirectional mirrors consisting of dielectric layers do exist [59, 60, 80]. The occurrence of omnidirectional reflection is promoted by the fact that a single dielectric layer and a multilayer stack both reflect  $p$ -polarized light at the Brewster angle [55]. This is to be contrasted with our plane scatterer model where the Brewster angle is shifted away to  $90^\circ$ . Therefore, the Brewster effect does help slabs but does not help planes to create the omnidirectional reflector.

What use can plane scatterers be in the future? One obvious idea is to study crystals built up of non-parallel planes. This turns out to be problematic, however, not so much

because the planes have an overlap of measure zero, but rather because the scattering of each plane as a whole must be described in its own plane representation. For that reason one can not treat the individual planes as wholes. Instead, one is forced to think of the planes as being built up of infinitely many line scatterers or point scatterers.

Albeit for parallel planes, the plane-scattering formalism can be used to study much more than was presented here. With the knowledge of the complete Green function, one could calculate near-field and far-field spectra. One could study the effect of more than one plane in a unit cell. Furthermore, the planes need not be identical and in fact the result (3.44) for the T-matrix also holds for planes chosen at arbitrary positions with different effective thicknesses. The model can therefore be used to study the effect of disorder, both in the positions and in the effective thicknesses of individual planes. Disorder can be increased further to study random dielectrics rather than crystals of plane scatterers.

Crystals of plane scatterers can also be used as a model system to study the modification of several quantum optical processes of embedded atoms. Transient effects in the spontaneous-emission rates are a first example, thereby generalizing work done on a one-dimensional cavity formed by two planes [81]. It is also interesting to study (two-atom) superradiance modified by the crystal (see chapter 5). Or one could generalize the formalism by allowing light absorption or gain in the planes, by choosing the effective thickness complex and frequency-dependent. In that case, a quantum optical description would require the identification of quantum noise operators (see chapter 6) for the planes. Also, Casimir forces can be calculated between an arbitrary number of imperfect mirrors. For two passive mirrors in one dimension, these forces are always attractive [82]. For planes with optical gain, the Casimir force might change sign.

## Chapter 4

# Multipole interaction between atoms and their photonic environment

*Macroscopic field quantization is presented for inhomogeneous nondispersive dielectrics, both with and without guest atoms. It is studied in detail how to obtain Maxwell's equations after choosing a suitable gauge. A quantum multipolar Hamiltonian is derived, starting from a classical minimal-coupling Lagrangian. Both photonic and electronic interactions of the atoms are discussed, in particular the dipole coupling of the atoms with the field.*

### 4.1 Introduction

In this chapter it will be studied how guest atoms interact with a photonic dielectric environment that is characterized by a given spatially varying and real dielectric function  $\varepsilon(\mathbf{r})$ . A quantum mechanical description will be presented in which the guest atoms will be described microscopically, whereas the dielectric is described macroscopically. This hybrid description is more economical and probably gives more understanding of photonic effects in inhomogeneous dielectrics than a more involved theory would do in which the dielectric is also described microscopically.

The quantization of the electromagnetic field in vacuum can be found in many textbooks on quantum optics [15, 45]. In [10, 11] the more general problem is addressed how to quantize the electromagnetic field in a dielectric described by a real dielectric function  $\varepsilon(\mathbf{r})$  that depends on position. The term “macroscopic quantization” has been coined for this procedure where the goal is not to derive a dielectric function from the interaction of light with the individual atoms that make up the dielectric [13]. Instead, the dielectric function is a given quantity. Material dispersion and absorption (transitions to nonradiative states in the dielectrics) are neglected. Certain sum rules [79] for modified spontaneous

emission rates when averaged over all frequencies will therefore not hold in the present formalism. Admittedly, the dielectric function is a response function that should be a dispersive and complex function of frequency. However, in the finite frequency interval of our interest, the dielectric function can be taken nondispersive and real. The same approximation is made for example when a mirror is assumed to be perfect, or in the chapters 2 and 3 of this thesis where the effective thicknesses of the plane scatterers were assumed real and frequency-independent. Also, in most band-structure calculations for photonic crystals a frequency-independent refractive-index contrast is assumed.

One can furthermore think of glass plates, optical cavities or dielectric mirrors as the type of dielectrics that can be characterized by a position-dependent dielectric function or refractive index. In these cases, the refractive index can often be considered as piecewise constant. Multimode glass fibres used in optical telecommunication do not have piecewise constant refractive indices: in the so-called graded-index fibres the refractive index in the core varies parabolically with the radius [83]. In this chapter, the relative dielectric function  $\varepsilon(\mathbf{r})$  is left unspecified (but assume it to be piecewise continuously differentiable) so that the theory describes dielectrics in both of the situations just sketched.

This rather formal chapter generalizes standard electrodynamics in the Coulomb gauge [84] to inhomogeneous dielectrics. The results are used in chapter 5 in order to derive a point-scattering formalism for quantum optics. The present chapter consists of two parts: the first part starts with the quantization of the electromagnetic field in inhomogeneous dielectrics without guest atoms. In the second and major part, the guest atoms are added and a Lagrangian is introduced with minimal-coupling interaction between field and atoms. In the end a quantum Hamiltonian is obtained with multipolar interaction between light and matter. In particular it is found to which field a dipole couples in a quantum optical description of inhomogeneous dielectrics. Even for an atom in free space this has been a controversial subject. In the discussion at the end of the chapter it is explained why controversy and confusion could arise; the free-space case is compared to the more general situation that is presented here of an atom in an inhomogeneous dielectric.

## 4.2 Inhomogeneous dielectric without guest atoms

### 4.2.1 Classical Lagrangian and Hamiltonian

In this section the quantization of the electromagnetic field in inhomogeneous dielectrics is briefly reviewed. The goal is to simplify derivations of the original texts [10, 11] and to derive results that will be useful in the following sections, when guest atoms are added in the dielectric.

In SI-units, the source-free Maxwell equations in matter are

$$\begin{aligned}
 \nabla \cdot \mathbf{D} &= 0 \\
 \nabla \cdot \mathbf{B} &= 0 \\
 \nabla \times \mathbf{H} - \dot{\mathbf{D}} &= 0 \\
 \nabla \times \mathbf{E} + \dot{\mathbf{B}} &= 0,
 \end{aligned} \tag{4.1}$$

where here and in the following the dot denotes a partial time derivative. The fields  $\mathbf{E}$ ,  $\mathbf{B}$ ,  $\mathbf{D}$  and  $\mathbf{H}$  are the electric field and the magnetic induction, the displacement field and the magnetic field vector. For nonmagnetic inhomogeneous dielectrics, the constitutive relations are simply  $\mathbf{B} = \mu_0 \mathbf{H}$  and  $\mathbf{D} = \varepsilon_0 \varepsilon(\mathbf{r}) \mathbf{E}$ . (The generalization of the present theory to inhomogeneous magnetic materials will be discussed at the end of this chapter in section 4.5.) As in the case of vacuum, the electric and magnetic fields can be expressed in terms of a vector potential  $\mathbf{A}$  and a scalar potential  $\Phi$ :

$$\mathbf{E} = -\nabla\Phi - \dot{\mathbf{A}} \quad (4.2)$$

$$\mathbf{B} = \nabla \times \mathbf{A} \quad (4.3)$$

There is gauge freedom in choosing pairs  $(\mathbf{A}, \Phi)$  that lead to the same electric and magnetic fields. Now choose the generalized Coulomb gauge which is defined by the requirement that the vector potential satisfies

$$\nabla \cdot [\varepsilon(\mathbf{r}) \mathbf{A}(\mathbf{r})] = 0. \quad (4.4)$$

The vector potential or any field satisfying this condition, is called “generalized transverse”, because it satisfies a generalized version of the Coulomb gauge condition  $\nabla \cdot \mathbf{A} = 0$  in free space. In this gauge, the vector potential must satisfy the wave equation

$$\nabla \times \nabla \times \mathbf{A} + \frac{\varepsilon(\mathbf{r})}{c^2} \ddot{\mathbf{A}} = 0, \quad (4.5)$$

in order to be consistent with the third Maxwell equation in (4.1). The scalar potential can be chosen identically zero in the generalized Coulomb gauge.

A Lagrangian  $L_0$  that leads to the wave equation (4.5) for the vector potential is

$$L_0 = \int d\mathbf{r} \mathcal{L}_0 \equiv \frac{1}{2} \int d\mathbf{r} \left[ \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{A}}^2 - \mu_0^{-1} (\nabla \times \mathbf{A})^2 \right]. \quad (4.6)$$

The Lagrangian is the spatial integral over the Lagrangian density  $\mathcal{L}_0$ . The vector potential is a canonical field variable and its canonically conjugate field can be found as a functional derivative of the Lagrangian density

$$\mathbf{\Pi}(\mathbf{r}, t) \equiv \frac{\delta L_0}{\delta \dot{\mathbf{A}}} = \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{A}} = -\mathbf{D}(\mathbf{r}, t). \quad (4.7)$$

(The functional derivative is used somewhat naively here, but the answer is correct, as a more detailed analysis in appendix B shows.) In other words, the field canonically conjugate to the vector potential equals minus the displacement field, which is a transverse field. The Hamiltonian is the integral over the Hamiltonian (energy) density  $\mathcal{H}$ :

$$H_0 = \int d\mathbf{r} \mathcal{H}_0 = \int d\mathbf{r} \left[ \mathbf{\Pi} \cdot \dot{\mathbf{A}} - \mathcal{L}_0 \right] = \frac{1}{2} \int d\mathbf{r} \left[ \frac{\mathbf{\Pi}^2}{\varepsilon_0 \varepsilon(\mathbf{r})} + \frac{(\nabla \times \mathbf{A})^2}{\mu_0} \right]. \quad (4.8)$$

This is the Hamiltonian for the classical electromagnetic field in an inhomogeneous dielectric, without the presence of guest atoms.

### 4.2.2 Complete sets and quantum Hamiltonian

For a quantum optical description of the dielectric, the electromagnetic fields can best be expanded in terms of harmonic solutions of the wave equation (4.5). With each of these “true modes” one can associate independent canonical variables, for which commutation rules can be given. The set of true modes is not unique. This freedom will be used below to choose a particularly convenient set. For example, in vacuum the true modes are plane waves. There one can choose linear combinations of cosine and sine solutions  $\cos(\mathbf{k} \cdot \mathbf{r})$  and  $\sin(\mathbf{k} \cdot \mathbf{r})$ . The complex exponential  $\exp(i\mathbf{k} \cdot \mathbf{r})$  is only one such linear combination.

Now a convenient set of true modes for the dielectric will be chosen and their orthonormality relations derived. Assume that the electromagnetic fields live in a large quantization volume  $V$  that can be chosen infinitely large at a later stage. Let  $Q$  be an abstract operator in Hilbert space which has a local representation in real space:

$$\langle \mathbf{r} | Q | \mathbf{r}' \rangle = \delta(\mathbf{r} - \mathbf{r}') Q(\mathbf{r}), \quad \text{with} \quad Q(\mathbf{r}) \equiv \frac{1}{\sqrt{\varepsilon(\mathbf{r})}} \nabla \times \nabla \times \frac{1}{\sqrt{\varepsilon(\mathbf{r})}}. \quad (4.9)$$

The operator  $Q$  is Hermitian under the normal inner product. All eigenfunctions  $\mathbf{g}_\lambda$  of  $Q$  have the property  $\nabla \cdot [\sqrt{\varepsilon(\mathbf{r})} \mathbf{g}(\mathbf{r})] = 0$ . The subspace of functions in Hilbert space with the same transversality property is spanned by the eigenfunctions of  $Q$ . Now let  $C$  be the operator which is also local in real space and whose action in real space is to take the complex conjugate. The dielectric function in this context must also be viewed as an abstract operator  $\varepsilon$  with local representation in real space:  $\langle \mathbf{r} | \varepsilon | \mathbf{r}' \rangle = \delta(\mathbf{r} - \mathbf{r}') \varepsilon(\mathbf{r})$  [68]. The real-space representations of  $Q$  and  $C$  commute, because  $\varepsilon(\mathbf{r})$  is real. Then  $Q$  and  $C$  commute in any representation. From the fact that  $Q$  and  $C$  commute it follows that an orthonormal basis of real eigenfunctions  $\{\mathbf{g}_\lambda\}$  of  $Q$  can be chosen to span the subspace (with complex coefficients).

As a consequence, the vector potential  $\mathbf{A}$  can be expanded in terms of a complete set of *real* vector mode functions  $\{\mathbf{h}_\lambda(\mathbf{r})\} \equiv \{\sqrt{\varepsilon(\mathbf{r})} \mathbf{g}_\lambda(\mathbf{r})\}$ , which are the harmonic solutions of the wave equation (4.5):

$$-\nabla \times \nabla \times \mathbf{h}_\lambda(\mathbf{r}) + \frac{\varepsilon(\mathbf{r}) \omega_\lambda^2}{c^2} \mathbf{h}_\lambda(\mathbf{r}) = 0. \quad (4.10)$$

These mode functions satisfy the same generalized transversality condition (4.4) as the vector potential. The choice of this particular set of mode functions  $\{\mathbf{h}_\lambda\}$  simplifies the quantization of the electromagnetic field, as we will see. (Real mode functions were also used in [10], without the above motivation.) The functions are labelled with  $\lambda$ , where  $\lambda$  is understood to count both continua and discrete sets of solutions. As is clear from Eq. (4.10), unlike the  $\mathbf{g}_\lambda$  the functions  $\mathbf{h}_\lambda$  do not satisfy a Hermitian eigenvalue equation. From the orthonormality of the  $\mathbf{g}_\lambda$  it follows that the functions  $\mathbf{h}_\lambda(\mathbf{r})$  satisfy the generalized orthonormality condition

$$\int d\mathbf{r} \, \varepsilon(\mathbf{r}) \mathbf{h}_\lambda^*(\mathbf{r}) \cdot \mathbf{h}_{\lambda'}(\mathbf{r}) = \delta_{\lambda\lambda'}. \quad (4.11)$$

The complex-conjugation symbol  $*$  was written for future reference, since of course  $\mathbf{h}_\lambda$  is real. The integration volume of this integral is the quantization volume  $V$ . The equation

(4.11) will be called a generalized inner product of the modes  $\mathbf{h}_\lambda$  and  $\mathbf{h}_{\lambda'}$ . The mode functions scale as  $(V)^{-1/2}$ .

In a scattering situation, where  $\varepsilon(\mathbf{r})$  is a space-filling dielectric function plus a local modification within a scattering volume  $V_s$ , the contribution of the scattering volume to the integral (4.11) scales as  $V_s/V$ , which becomes of measure zero when the quantization volume  $V$  is sent to infinity. As an example, mode functions of a photonic crystal with a point defect have the same orthonormality relations as the other mode functions in the absence of the defect.

The functions  $\mathbf{h}_\lambda$  are complete in the sense that they form a basis for generalized transverse functions such as the vector potential that satisfy the wave equation (4.5). In other words, a generalized transverse delta function  $\delta_\varepsilon^T$  (a distribution) can be defined in terms of the functions  $\mathbf{h}_\lambda$ :

$$\delta_\varepsilon^T(\mathbf{r}, \mathbf{r}') \equiv \sum_\lambda \mathbf{h}_\lambda(\mathbf{r}) \mathbf{h}_\lambda(\mathbf{r}') \varepsilon(\mathbf{r}'). \quad (4.12)$$

For  $\varepsilon(\mathbf{r}) \equiv 1$ , this expression reduces to the free-space transverse delta function that will be given explicitly in Eq. (C.5) of appendix C, and which can be written as a dyadic sum over transverse plane wave modes (see [85], p. 53). Evidently, the generalized transverse delta function (4.12) is real because the mode functions are real. The “identity” property (4.12) together with the idempotence property

$$\int d\mathbf{r}_1 \delta_\varepsilon^T(\mathbf{r}, \mathbf{r}_1) \cdot \delta_\varepsilon^T(\mathbf{r}_1, \mathbf{r}') = \delta_\varepsilon^T(\mathbf{r}, \mathbf{r}'), \quad (4.13)$$

define  $\delta_\varepsilon^T$  as a projector in the subspace of generalized transverse functions. The generalized transverse delta function is not symmetric in its arguments, because it is transverse in its second and generalized transverse in its first variable:

$$\nabla_{\mathbf{r}} \cdot [\varepsilon(\mathbf{r}) \delta_\varepsilon^T(\mathbf{r}, \mathbf{r}')] = 0 = \nabla_{\mathbf{r}'} \cdot [\delta_\varepsilon^T(\mathbf{r}, \mathbf{r}')]. \quad (4.14)$$

Furthermore, it has the properties that

$$\mathbf{X}^T(\mathbf{r}) - \int d\mathbf{r}' \mathbf{X}^T(\mathbf{r}') \cdot \delta_\varepsilon^T(\mathbf{r}', \mathbf{r}) = 0, \quad (4.15a)$$

$$\mathbf{X}^T(\mathbf{r}) - \varepsilon(\mathbf{r}) \int d\mathbf{r}' \delta_\varepsilon^T(\mathbf{r}, \mathbf{r}') \cdot \mathbf{X}^T(\mathbf{r}') / \varepsilon(\mathbf{r}') = 0, \quad (4.15b)$$

$$\int d\mathbf{r}' \delta_\varepsilon^T(\mathbf{r}, \mathbf{r}') \cdot \mathbf{X}^L(\mathbf{r}') = 0, \quad (4.15c)$$

$$\int d\mathbf{r}' \varepsilon(\mathbf{r}') \mathbf{X}^L(\mathbf{r}') \cdot \delta_\varepsilon^T(\mathbf{r}', \mathbf{r}) = 0, \quad (4.15d)$$

for any transverse function  $\mathbf{X}^T$  (zero divergence) and longitudinal function  $\mathbf{X}^L$  (zero curl). These properties follow immediately from the transversality property (4.4) and the orthogonality relations (4.11) of the mode functions  $\mathbf{h}_\lambda$ . These several properties of the generalized transverse delta function will be used in later sections and in appendix B.

The vector potential and its canonically conjugate field have normal-mode expansions

$$\mathbf{A}(\mathbf{r}, t) = 1/(\sqrt{\varepsilon_0}) \sum_{\lambda} q_{\lambda}(t) \mathbf{h}_{\lambda}(\mathbf{r}), \quad (4.16a)$$

$$\mathbf{\Pi}(\mathbf{r}, t) = \sqrt{\varepsilon_0} \sum_{\lambda} p_{\lambda}(t) \varepsilon(\mathbf{r}) \mathbf{h}_{\lambda}(\mathbf{r}). \quad (4.16b)$$

If the above two expansions are substituted in the Hamiltonian (4.8) and the normalization condition (4.11) is used, then it follows that

$$H_0 = \frac{1}{2} \sum_{\lambda} (p_{\lambda}^2 + \omega_{\lambda}^2 q_{\lambda}^2). \quad (4.17)$$

The Hamiltonian turns out to be a simple sum over the true modes of the inhomogeneous dielectric, where the energy of each mode corresponds to a one-dimensional harmonic oscillator with position  $q_{\lambda}$ , momentum  $p_{\lambda}$  and frequency  $\omega_{\lambda}$ . This is a generalization to inhomogeneous dielectrics of the result for free space.

Now comes the quantization step. Classically, the generalized coordinates  $q_{\lambda}(t)$  and momenta  $p_{\lambda}(t)$  are real variables because  $\mathbf{A}(\mathbf{r}, t)$  and  $\mathbf{\Pi}(\mathbf{r}, t)$  and the modes  $\mathbf{h}_{\lambda}$  are real. Real variables such as  $q_{\lambda}(t)$  and  $p_{\lambda}(t)$  turn into Hermitian operators in quantum mechanics. The independent canonical pairs satisfy the standard equal-time commutation relations

$$[q_{\lambda}(t), p_{\lambda'}(t)] = i\hbar \delta_{\lambda\lambda'}. \quad (4.18)$$

With the normal mode expansions (4.16a) and (4.16b), the commutation relation for the vector potential and its canonically conjugate field can be found immediately:

$$[\mathbf{A}(\mathbf{r}, t), \mathbf{\Pi}(\mathbf{r}', t)] = i\hbar \sum_{\lambda} \mathbf{h}_{\lambda}(\mathbf{r}) \mathbf{h}_{\lambda}(\mathbf{r}') \varepsilon(\mathbf{r}') = i\hbar \delta_{\varepsilon}^T(\mathbf{r}, \mathbf{r}'). \quad (4.19)$$

The commutator is a dyadic quantity which turns out to be proportional to the generalized transverse delta function.

Creation and annihilation operators are introduced as

$$\alpha_{\lambda}(t) = \sqrt{\frac{\omega_{\lambda}}{2\hbar}} q_{\lambda}(t) + i\sqrt{\frac{1}{2\hbar\omega_{\lambda}}} p_{\lambda}(t) \quad (4.20a)$$

$$\alpha_{\lambda}^{\dagger}(t) = \sqrt{\frac{\omega_{\lambda}}{2\hbar}} q_{\lambda}(t) - i\sqrt{\frac{1}{2\hbar\omega_{\lambda}}} p_{\lambda}(t). \quad (4.20b)$$

They have standard commutation relations  $[\alpha_{\lambda}(t), \alpha_{\lambda'}(t)^{\dagger}] = \delta_{\lambda\lambda'}$  and all other inequivalent commutators are zero. The Hamiltonian becomes the sum over contributions of individual modes,  $\hbar\omega_{\lambda}(\alpha_{\lambda}^{\dagger}\alpha_{\lambda} + 1/2)$ . Thus the concept of a photon as the elementary excitation  $\alpha_{\lambda}^{\dagger}|0\rangle$  of a mode is as useful for inhomogeneous dielectrics as it is for free space. Number states, coherent and squeezed states etcetera can be defined analogously as for free space. The only difference for inhomogeneous dielectrics is that their true

modes are not plane waves. The vector potential and its canonically conjugate field can be expressed in terms of creation and annihilation operators as

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\lambda} \sqrt{\frac{\hbar}{2\varepsilon_0\omega_{\lambda}}} \left[ \alpha_{\lambda}(t) \mathbf{h}_{\lambda}(\mathbf{r}) + \alpha_{\lambda}^{\dagger}(t) \mathbf{h}_{\lambda}(\mathbf{r}) \right], \quad (4.21)$$

$$\mathbf{\Pi}(\mathbf{r}, t) = -i\varepsilon_0\varepsilon(\mathbf{r}) \sum_{\lambda} \sqrt{\frac{\hbar\omega_{\lambda}}{2\varepsilon_0}} \left[ \alpha_{\lambda}(t) \mathbf{h}_{\lambda}(\mathbf{r}) - \alpha_{\lambda}^{\dagger}(t) \mathbf{h}_{\lambda}(\mathbf{r}) \right]. \quad (4.22)$$

The forms of the electric and magnetic fields as quantum mechanical operators and their commutation relations immediately follow from (4.2), (4.3) and the above equation (4.21). The time-dependence of the operators is simply harmonic,  $\alpha_{\lambda}(t)$  equals  $\alpha(0) \exp(-i\omega_{\lambda}t)$ , for example. The quantization of the electromagnetic field has thus been accomplished.

In practice, it can be convenient to use a set of complex true mode functions  $\{\mathbf{f}_{\mu}\}$  instead of the real mode functions  $\{\mathbf{h}_{\lambda}\}$ . Since the complex mode functions should also satisfy the wave equation (4.10) and the generalized orthonormality condition (4.11), the two sets of mode functions are related through a unitary transformation that only relates mode function with identical eigenfrequencies:

$$\mathbf{f}_{\mu} = \sum_{\lambda} U_{\mu\lambda}^* \mathbf{h}_{\lambda}, \quad (4.23)$$

where  $\mathbf{U}^* = \mathbf{U}^{-1}$ . Note that because of the relation (4.23), the generalized transverse delta function (4.12) can alternatively be expressed in terms of the complex mode functions  $\mathbf{f}_{\lambda}$ . Its effect is the substitution of one  $\mathbf{h}_{\lambda}$  by  $\mathbf{f}_{\lambda}$  and the other  $\mathbf{h}_{\lambda}$  by  $\mathbf{f}_{\lambda}^*$ :

$$\delta_{\varepsilon}^T(\mathbf{r}, \mathbf{r}') = \sum_{\lambda} \mathbf{f}_{\lambda}(\mathbf{r}) \mathbf{f}_{\lambda}^*(\mathbf{r}') \varepsilon(\mathbf{r}'). \quad (4.24)$$

However, after this substitution it is no longer obvious that  $\delta_{\varepsilon}^T(\mathbf{r}, \mathbf{r}')$  is real-valued.

The electromagnetic fields can also be expanded in terms of the complex mode functions  $\mathbf{f}_{\lambda}$  as

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\lambda} \sqrt{\frac{\hbar}{2\varepsilon_0\omega_{\lambda}}} \left[ a_{\lambda}^{(0)}(t) \mathbf{f}_{\lambda}(\mathbf{r}) + a_{\lambda}^{(0)\dagger}(t) \mathbf{f}_{\lambda}^*(\mathbf{r}) \right], \quad (4.25a)$$

$$\mathbf{\Pi}(\mathbf{r}, t) = -i\varepsilon_0\varepsilon(\mathbf{r}) \sum_{\lambda} \sqrt{\frac{\hbar\omega_{\lambda}}{2\varepsilon_0}} \left[ a_{\lambda}^{(0)}(t) \mathbf{f}_{\lambda}(\mathbf{r}) - a_{\lambda}^{(0)\dagger}(t) \mathbf{f}_{\lambda}^*(\mathbf{r}) \right], \quad (4.25b)$$

where the new annihilation operator  $a_{\mu}^{(0)}$  associated with the complex mode  $\mathbf{f}_{\mu}$  is defined in terms of the “old” operators as

$$a_{\mu}^{(0)} \equiv \sum_{\lambda} U_{\mu\lambda} \alpha_{\lambda}, \quad (4.26)$$

and  $a_{\mu}^{(0)\dagger}$  is its Hermitian conjugate. The commutation relations of  $a_{\mu}^{(0)}$  and  $a_{\mu}^{(0)\dagger}$  are again the standard relations, because  $\mathbf{U}$  is a unitary transformation. To distinguish  $a_{\mu}^{(0)}$

from operators to be defined later, the superscript (0) has been added, signifying that no guest atoms are present. The time dependence of the operators is harmonic.

This completes the quantization of the electromagnetic field in an inhomogeneous dielectric without guest atoms. The reason not to start the quantization procedure with complex mode functions is that the associated generalized coordinates and momenta are not Hermitian. This makes intermediate results more complicated [11, 13, 86]. These unnecessary complications were avoided here. The relations (4.23) between complex and real mode functions and (4.26) between their respective annihilation operators are purely formal, unless both sets of mode functions are given explicitly. It is the mere existence of the mapping (4.23) of the complex modes onto the real modes that justifies the expansion of the operators  $\mathbf{A}$  and  $\mathbf{\Pi}$  in terms of the complex mode functions. The choice of real mode functions made explicit otherwise implicit properties of the delta function  $\delta_{\epsilon}^T$ .

### 4.3 Inhomogeneous dielectric with guest atoms

In the previous section it was described how to quantize the electromagnetic field in an inhomogeneous dielectric. Now inside the inhomogeneous dielectric guest atoms are introduced. Their optical response is not included in the dielectric function  $\epsilon(\mathbf{r})$  of the medium. The goal in the following sections is to find the quantum optical description of the combined system, with a multipole interaction between the electromagnetic field and the guest atoms. There are at least two reasons why the multipolar Hamiltonian is to be preferred. In the first place, it is more convenient when only approximate calculations can be done which in the minimal-coupling formalism would give gauge-dependent results [15]; secondly, atoms are much smaller than optical wavelengths and in the multipole-formalism this can be exploited well. Actually, atoms are so much smaller than optical wavelengths that often “atoms” are identified with “dipoles”.

The starting point is the minimal-coupling Lagrangian that produces the Maxwell equations and the equations of motion for the charges that make up the guest atoms. The minimal-coupling Lagrangian can be used to find a minimal-coupling Hamiltonian and this procedure can be found in [10, 87]. A clear exposition is also given in [88]. Here the Lagrangian will first be transformed to the multipolar form before constructing a Hamiltonian. The latter procedure was followed also in [13, 14]. The present work is different in that a careful treatment of functional differentiation is given and that a qualitative connection of final results with local-field effects will be made. Helpful details of the quantization of the vacuum electromagnetic field in the presence of atoms can be found in the books [15, 84, 86].

### 4.3.1 Choice of suitable Lagrangian

Guest atoms inside an inhomogeneous dielectric can be described by a charge density  $\sigma_g$  and a current density  $\mathbf{J}_g$  which show up as sources in Maxwell's equations:

$$\nabla \cdot \mathbf{B} = 0, \quad (4.27)$$

$$\nabla \times \mathbf{E} + \dot{\mathbf{B}} = 0, \quad (4.28)$$

$$\varepsilon_0 \nabla \cdot [\varepsilon(\mathbf{r}) \mathbf{E}(\mathbf{r})] = \sigma_g, \quad (4.29)$$

$$\mu_0^{-1} \nabla \times \mathbf{B} - \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{E}} = \mathbf{J}_g. \quad (4.30)$$

Whatever Lagrangians and Hamiltonians are introduced for the inhomogeneous dielectric plus guest atoms, they must lead to these four Maxwell equations. Moreover, the particles with charges  $q_{mj}$  and masses  $M_{mj}$  should respond to electric and magnetic fields as given in the equation of motion

$$M_{mj} \ddot{\mathbf{r}}_{mj} = q_{mj} [\mathbf{E}(\mathbf{r}_{mj}) + \dot{\mathbf{r}}_{mj} \times \mathbf{B}(\mathbf{r}_{mj})]. \quad (4.31)$$

We assume that there are no free charges. All electrons (labelled  $j$ ) are bound to atomic nuclei (label  $m$ ) to form neutral atoms that are constituent of the guest atoms. Let  $Z_m e$  be the nuclear charge of atom  $m$ . Then  $\sigma_g$  and  $\mathbf{J}_g$  are given by [4]

$$\sigma_g(\mathbf{r}) = \sum_m \left[ Z_m e \delta(\mathbf{r} - \mathbf{R}_m) - e \sum_j \delta(\mathbf{r} - \mathbf{r}_{mj}) \right], \quad (4.32)$$

$$\mathbf{J}_g(\mathbf{r}) = \sum_m \left[ -e \sum_j \dot{\mathbf{r}}_{mj} \delta(\mathbf{r} - \mathbf{r}_{mj}) \right]. \quad (4.33)$$

These are charge and current densities of the guest atoms alone, as stressed by the subscript  $g$ ; the dielectric is completely described by the dielectric function  $\varepsilon_0 \varepsilon(\mathbf{r})$  and the magnetic permeability  $\mu_0$ . The guest atoms are assumed to have fixed positions, their nuclei are their centers of mass and are stationary at positions  $\mathbf{R}_m$ . From these explicit forms of  $\sigma_g$  and  $\mathbf{J}_g$  or from the third and fourth Maxwell equation follows the equation of continuity or current conservation, namely

$$\nabla \cdot \mathbf{J}_g + \dot{\sigma}_g = 0. \quad (4.34)$$

Again, the electric and magnetic fields can be defined through Eq (4.2) and (4.3) in terms of a vector potential  $\mathbf{A}$  and a scalar potential  $\Phi$ . Then the first two Maxwell equations are automatically satisfied. The third and fourth Maxwell equations should follow from the Euler-Lagrange equations for the scalar and the vector potential, respectively. The minimal-coupling Lagrangian is

$$L_{\min} = \sum_m \left[ \sum_j \frac{1}{2} m_e \dot{\mathbf{r}}_{mj}^2 \right] + \int d\mathbf{r} \mathcal{L}_{\min}. \quad (4.35)$$

Here the Lagrangian density  $\mathcal{L}_{\min}$  describes the electromagnetic field energy and its minimal-coupling interaction with the guest atoms:

$$\mathcal{L}_{\min} = \frac{1}{2}\varepsilon_0\varepsilon(\mathbf{r}) \left[ \dot{\mathbf{A}} + \nabla\Phi \right]^2 - \frac{1}{2\mu_0}(\nabla \times \mathbf{A})^2 + \mathbf{J}_g \cdot \mathbf{A} - \sigma_g\Phi. \quad (4.36)$$

Indeed, Maxwell's third and fourth equations can be found from the Euler-Lagrange equations for the scalar and the vector potentials, respectively. Moreover, the Euler-Lagrange equations for the canonical variables  $\mathbf{r}_{mj}$  give the equations of motion (4.31) for the charged particles. Note that the Lagrangian leads to these equations of motion, before choosing a gauge to fix  $\mathbf{A}$  and  $\Phi$  with: the equations of motion are gauge-independent results that should not depend on the choice of gauge.

### 4.3.2 Fixing the gauge

The electric and magnetic fields are defined in terms of a scalar and a vector potential. But there is gauge freedom, which means that the scalar and vector potentials are not uniquely defined by the requirement that physical measurable electric and magnetic fields satisfy Maxwell's equations. We need to choose a gauge in order to find in the end a quantum mechanical description of light interacting with the guest atoms. It is not necessary to choose the gauge already at this point, but is a matter of presentation to do so. In the conclusions in section 4.7 more will be said about this. As in the situation without guest atoms in section 4.2, the generalized Coulomb gauge is chosen in which  $\nabla \cdot [\varepsilon(\mathbf{r})\mathbf{A}(\mathbf{r})] = 0$ . In this section it will be checked whether the equations of motion for the scalar and vector potentials still lead to the third and fourth Maxwell equations after choosing the generalized Coulomb gauge. This must be the case, because the choice of gauge should not change the physical predictions of the theory.

The interaction term

$$L_{\mathbf{A}\Phi} \equiv \int d\mathbf{r} \varepsilon_0\varepsilon(\mathbf{r})\dot{\mathbf{A}} \cdot \nabla\Phi, \quad (4.37)$$

of the Lagrangian (4.35) becomes identically zero, because in the generalized Coulomb gauge it has become an inner product of a transverse and a longitudinal function. The remaining terms in the Lagrangian involving the scalar potential lead to an Euler-Lagrange equation that is the generalized Poisson equation for the scalar potential in the Coulomb gauge:

$$\varepsilon_0\nabla \cdot [\varepsilon(\mathbf{r})\nabla\Phi(\mathbf{r})] = -\sigma_g. \quad (4.38)$$

Clearly, the scalar potential can not be chosen identically zero as in the situation without guest atoms. The gauge-fixing condition (4.4) for the vector potential, the equation (4.38) for the scalar potential, together with the definition of the electric field (4.2) in terms of the two potentials, still lead to the third Maxwell equation (4.29). This is as it should be, since Maxwell's equations should not change with the choice of the gauge.

The scalar potential is a function of the positions of the charges that make up the guest atoms. In other words, one can first solve the coupled equations of motion for the vector potential and the charges, and from the charge distribution  $\sigma_g(t)$  thus found,

the scalar potential  $\Phi(t)$  can be found as the solution of Eq. (4.38). Therefore, in the generalized Coulomb gauge the scalar potential is not an independent canonical field. Then the Lagrangian (4.35) can be simplified as

$$L_{\min} = \sum_m \left[ \sum_j \frac{1}{2} m_e \dot{\mathbf{r}}_{mj}^2 \right] - V_C + \int d\mathbf{r} \mathcal{L}'_{\min}, \quad (4.39)$$

where Eqs. (4.37) and (4.38) were used. The Coulomb interaction

$$V_C = \frac{\varepsilon_0}{2} \int d\mathbf{r} \varepsilon(\mathbf{r}) (\nabla \Phi)^2 \quad (4.40)$$

is a function of the guest atoms alone; the Lagrangian density in (4.39) becomes

$$\mathcal{L}'_{\min} = \frac{1}{2} \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{A}}^2 - \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 + \mathbf{J}_g \cdot \mathbf{A}. \quad (4.41)$$

Which equation do we find for the vector potential after choosing the generalized Coulomb gauge? Let us begin at the other end: in order to be consistent with the fourth Maxwell equation, the vector potential should satisfy

$$\mu_0^{-1} \nabla \times \nabla \times \mathbf{A} + \varepsilon_0 \varepsilon(\mathbf{r}) \ddot{\mathbf{A}} = \mathbf{J}_g - \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \dot{\Phi}. \quad (4.42)$$

It is not easy to see how the source term  $-\varepsilon_0 \varepsilon(\mathbf{r}) \nabla \dot{\Phi}$  can appear at the right-hand side of this equation by functional differentiation of the Lagrangian with respect to the vector potential. Before choosing the gauge, the source term originated from the interaction term  $L_{\mathbf{A}\Phi}$  [Eq. (4.37)], which is zero after choosing the gauge. Functional derivatives of zero should of course be zero. At this point, it seems hard to find the fourth Maxwell equation after choosing the generalized Coulomb gauge.

In appendix B it is explained in detail that the usual functional derivative must be replaced by a “constrained functional derivative” after choosing a gauge. This is for mathematical reasons rather than a matter of taste or convenience. In the appendix one also finds simple rules to actually compute these constrained functional derivatives. In particular, it is shown that the Euler-Lagrange equation for the vector potential indeed gives (4.42) when the correct functional derivative is used. Interestingly, before choosing the gauge, the source term  $-\varepsilon_0 \varepsilon(\mathbf{r}) \nabla \dot{\Phi}$  in (4.42) came from the  $L_{\mathbf{A}\Phi}$  interaction term (4.37) in the Lagrangian. However, after choosing the gauge the source term is produced by the constrained functional derivative of the minimal-coupling interaction term  $\int d\mathbf{r} \mathbf{J}_g \cdot \mathbf{A}$ .

The left-hand side of the wave equation (4.42) is certainly transverse in the generalized Coulomb gauge. It is less obvious and sometimes goes unnoticed [13, 88] that the source term on the right-hand side is also transverse. A mathematical reason is that the wave equation is found by functional differentiation with respect to generalized transverse functions. In section B.2 it was shown that these derivatives are always transverse. Physically, the source term must be transverse because of current conservation, Eq. (4.34). In [88] it was shown that the vector potential satisfying the wave equation (4.42) can not be influenced by the longitudinal part of the source term. Here one must conclude that in the generalized Coulomb gauge a longitudinal part of the source term simply does not exist.

## 4.4 The quantum multipolar interaction Hamiltonian

In the previous sections it was shown that the minimal-coupling Lagrangian produces the Maxwell-Lorentz equations for the electromagnetic fields and the guest charges, before and also after choosing the generalized Coulomb gauge. Now the goal is to transform the Lagrangian in order to obtain a multipole interaction between the electromagnetic field and the guest atoms. The transformed Lagrangian must lead to the same equations for the fields and charges, of course. Candidate transformations are transformations where a total time derivative of a function of the canonical variables is added to the Lagrangian [84]. Such a transformation will be used shortly, but first some new fields must be introduced.

### 4.4.1 Polarization, magnetization and displacement fields

In the following, it is useful to describe the guest atoms in terms of a polarization density  $\mathbf{P}_g$  and a magnetization density  $\mathbf{M}_g$ , rather than in terms of the charge and current densities. The former and latter pairs are related through [4]:

$$\sigma_g = -\nabla \cdot \mathbf{P}_g, \quad \mathbf{J}_g = \dot{\mathbf{P}}_g + \nabla \times \mathbf{M}_g. \quad (4.43)$$

In terms of the new variables, the equation of continuity (4.34) is automatically satisfied. By using the definitions (4.32) and (4.33) of  $\sigma_g$  and  $\mathbf{J}_g$  and by making a Taylor expansion around the atomic positions  $\mathbf{R}_m$  in both the left-hand and the right-hand sides of the above definitions, one can find that

$$\mathbf{P}_g(\mathbf{r}) = -e \sum_{mj} \int_0^1 du (\mathbf{r}_{mj} - \mathbf{R}_m) \delta(\mathbf{r} - \mathbf{R}_m - u(\mathbf{r}_{mj} - \mathbf{R}_m)), \quad (4.44)$$

$$\mathbf{M}_g(\mathbf{r}) = -e \sum_{mj} \int_0^1 du u (\mathbf{r}_{mj} - \mathbf{R}_m) \times \dot{\mathbf{r}}_{mj} \delta(\mathbf{r} - \mathbf{R}_m - u(\mathbf{r}_{mj} - \mathbf{R}_m)). \quad (4.45)$$

Finite-order multipole expansions of the polarization and magnetization fields can be found by truncating the Taylor expansion in  $u$  of the integrand on the right-hand sides of the above equations. Such approximations will be made later, in section 4.4.5.

The displacement field  $\mathbf{D}$  and the magnetic field vector  $\mathbf{H}$  are given by the constitutive relations

$$\mathbf{D} \equiv \varepsilon_0 \varepsilon(\mathbf{r}) \mathbf{E} + \mathbf{P}_g \quad \mathbf{H} \equiv \mu_0^{-1} \mathbf{B} - \mathbf{M}_g, \quad (4.46)$$

where in the latter relation it was assumed as before that the dielectric is nonmagnetic so that the magnetic permeability equals the value  $\mu_0$  of free space. Note that the dielectric function  $\varepsilon(\mathbf{r})$  is a property of the dielectric alone, independent of the guest atoms. On the other hand, the displacement field (4.46) does depend on the guest atoms because it includes the polarization field produced by them. The displacement field defined here is therefore different (although the same symbol is used) from the displacement field that was defined in section 4.2.1 where no guest atoms were present. In that case, the transversality of  $\mathbf{D}$  was evident. The displacement field is also transverse when guest atoms are present, according to eqs. (4.29) and (4.43) in combination with (4.46).

### 4.4.2 Classical multipolar Lagrangian and Hamiltonian

The Lagrangian will be transformed by adding to it the total time derivative

$$-\frac{d}{dt} \int d\mathbf{r} \mathbf{P}_g(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}). \quad (4.47)$$

This is the Power-Zienau-Woolley (PZW) transformation, and its effect is well-known for free space [84]. The PZW transformation was already applied to inhomogeneous dielectrics before, in [13]. There it was stated that the polarization density  $\mathbf{P}_g$  in the Lagrangian density  $\mathcal{L}'$  should be replaced by a “reduced polarization density” at this point, in order to stick to the generalized Coulomb gauge for the vector potential. However, such replacements are not necessary if functional derivatives with respect to the generalized transverse vector potential are identified as constrained functional differentiations, and if computation rules are used accordingly (see appendix B).

After adding the term (4.47), the new Lagrangian  $L_{\text{multi}}$  can be rewritten as

$$L_{\text{multi}} = \sum_{mj} \frac{1}{2} m_e \dot{\mathbf{r}}_{mj}^2 - V_C + \int d\mathbf{r} \mathcal{L}_{\text{multi}}. \quad (4.48)$$

The new Lagrangian density  $\mathcal{L}_{\text{multi}}$  has the form

$$\mathcal{L}_{\text{multi}} = \frac{1}{2} \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{A}}^2(\mathbf{r}) - \frac{1}{2\mu_0} [\nabla \times \mathbf{A}(\mathbf{r})]^2 + \mathbf{M}_g(\mathbf{r}) \cdot \nabla \times \mathbf{A}(\mathbf{r}) - \mathbf{P}_g(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}), \quad (4.49)$$

where the definition of the magnetization density in (4.44) was used as well as Gauss’s theorem. The derivation is identical to the free-space case.

In order to find a Hamiltonian, first the canonically conjugate variables must be determined. By reasoning as in section B.3, the constrained functional differentiation of the Lagrangian  $L_{\text{multi}}$  with respect to  $\dot{\mathbf{A}}$  produces the following field that is canonically conjugate to the vector potential:

$$\Pi(\mathbf{r}, t) \equiv \frac{\delta L_{\text{multi}}}{\delta \dot{\mathbf{A}}_e^T(\mathbf{r})} = \varepsilon_0 \varepsilon(\mathbf{r}) \dot{\mathbf{A}}(\mathbf{r}) - \mathbf{P}_g(\mathbf{r}) + \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \Phi(\mathbf{r}) = -\mathbf{D}(\mathbf{r}). \quad (4.50)$$

In the latter identity the definition of the electric field (4.2) and the displacement field (4.46) were used. As in the case without guest atoms, the field canonically conjugate to the vector potential equals minus the displacement field. The difference is that now the displacement field also contains the polarization field produced by the guest atoms. The canonically conjugate field would have been different if the minimal-coupling Lagrangian had been used.

The canonical momenta  $\mathbf{p}_{mj}$  corresponding to the coordinate variables  $\mathbf{q}_{mj}$  of the guest charges are

$$\mathbf{p}_{mj} = m_e \dot{\mathbf{r}}_{mj} - \mathbf{F}_{mj}, \quad (4.51)$$

where the field  $\mathbf{F}_{mj}$  stems from the magnetization density in Eq. (4.44) and is defined as

$$\mathbf{F}_{mj} \equiv e \int_0^1 du \, u \, \mathbf{B}[\mathbf{R}_m - u (\mathbf{r}_{mj} - \mathbf{R}_m)] \times (\mathbf{r}_{mj} - \mathbf{R}_m). \quad (4.52)$$

The magnetic field  $\mathbf{B}$  is used as an abbreviation for  $\nabla \times \mathbf{A}$ . Note that unlike  $\mathbf{A}$  and  $\mathbf{q}_{mj}$ , their canonically conjugate variables  $\mathbf{\Pi}$  and  $\mathbf{p}_{mj}$  are not fully electromagnetic or fully atomic in nature, respectively.

All canonical momenta have now been determined, so that the multipolar Hamiltonian  $H'$  can be given in terms of the canonical variables  $(\mathbf{r}_{mj}, \mathbf{p}_{mj})$  and  $(\mathbf{A}, \mathbf{\Pi})$ :

$$\begin{aligned}
 H_{\text{multi}} &= \sum_{mj} \mathbf{p}_{mj} \cdot \dot{\mathbf{r}}_{mj} + \int d\mathbf{r} \, \mathbf{\Pi} \cdot \dot{\mathbf{A}} - L_{\text{multi}} \\
 &= \sum_{mj} \frac{p_{mj}^2}{2m_e} + \int d\mathbf{r} \, \frac{\mathbf{P}_g^2}{2\varepsilon_0\varepsilon(\mathbf{r})} + \int d\mathbf{r} \, \left[ \frac{\mathbf{\Pi}^2}{2\varepsilon_0\varepsilon(\mathbf{r})} + \frac{\mathbf{B}^2}{2\mu_0} \right] \\
 &\quad + \sum_{mj} \frac{F_{mj}^2}{2m_e} + \int d\mathbf{r} \, \left[ \frac{\mathbf{P}_g \cdot \mathbf{\Pi}}{\varepsilon_0\varepsilon(\mathbf{r})} - \mathbf{M}_g' \cdot \mathbf{B} \right].
 \end{aligned} \tag{4.53}$$

Instead of the magnetization  $\mathbf{M}_g$ , a reduced magnetization  $\mathbf{M}_g'$  has been used in the Hamiltonian (and the difference has been corrected for by a sign change of the term quadratic in the magnetic field). The reduced magnetization is defined as the magnetization [see equation (4.44)] with the  $\dot{\mathbf{r}}_{mj}$  replaced by  $\mathbf{p}_{mj}/m_e$  [13, 84]. It may seem that the Coulomb term  $V_C$  (4.40) was forgotten in the Hamiltonian, but that is not the case: the Coulomb term cancels against the other term quadratic in  $\nabla\Phi$  that one gets when solving Eq. (4.50) for  $\dot{\mathbf{A}}$  and substituting the result in the Hamiltonian (4.53).

The first term in the Hamiltonian (4.53) represents the kinetic energy of the guest charges; the second term is the potential energy of the guest atoms, expressed as a polarization energy. Together they constitute the atomic part of the Hamiltonian. Next come the electric and magnetic field energies, respectively, which together are the radiative part of the Hamiltonian. The last three terms are interaction terms between guest atoms and the electromagnetic field. The term quadratic in the magnetic field represents the diamagnetic energy of only the guest atoms in the nonmagnetic dielectric. It can be safely ignored from now on since it is much smaller than the other two interactions (see [15], page 312). In most cases the dominant interaction term is the next one, the inner product of the polarization field with the field  $\mathbf{\Pi}/[\varepsilon_0\varepsilon(\mathbf{r})]$ . The last term is linear in the magnetic field and represents the paramagnetic energy of the guest atoms in the dielectric.

The polarization field  $\mathbf{P}_g$  [Eq. (4.44)] is simply the sum of the polarization fields  $\mathbf{P}_{gm}$  produced by the individual guest atoms (labeled by  $m$ ). Therefore, it is natural to split the polarization energy in Eq. (4.53) into an intra-atomic and an interatomic polarization energy, respectively [14, 84]:

$$\int d\mathbf{r} \, \frac{\mathbf{P}_g^2}{2\varepsilon_0\varepsilon(\mathbf{r})} = \sum_m \int d\mathbf{r} \, \frac{P_{gm}^2}{2\varepsilon_0\varepsilon(\mathbf{r})} + \sum_{n \neq m} \int d\mathbf{r} \, \frac{\mathbf{P}_{gn} \cdot \mathbf{P}_{gm}}{\varepsilon_0\varepsilon(\mathbf{r})}. \tag{4.54}$$

The intra-atomic polarization energy is the potential energy that keeps an atom together; the interatomic polarization energy is the only interaction term between neutral atoms in the multipolar Hamiltonian. The polarization field  $\mathbf{P}_{gm}(\mathbf{r})$  (4.44) corresponding to the individual guest atom  $m$  is identically zero outside the smallest sphere surrounding all

charges that make up this (neutral) atom. In this classical picture, the interatomic polarization energy (also known as contact energy) is zero unless bounding spheres of distinct guest atoms overlap. Correspondingly, in quantum mechanics the atomic wave functions of distinct guest atoms have a nonvanishing overlap that falls off exponentially with interatomic distance. The overlap is therefore negligible unless the interatomic distance is of the order of the size of the atoms.

From now on the guest atoms are assumed more than a few nanometers apart so that the contact energy can be neglected. Just like in the free-space case [84], in the multipole Hamiltonian (4.53) for the inhomogeneous dielectric there is no instantaneous interaction term left between well-separated neutral guest atoms: the effect of the Coulomb term exactly cancelled against other terms. All other interatomic interactions are radiative interactions, mediated by the (retarded) electromagnetic fields  $\mathbf{D}/[\varepsilon_0\varepsilon(\mathbf{r})]$  and  $\mathbf{B}$ . Of course, the multipolar representation should give the same physical predictions as the minimal-coupling formalism; in [84, 85] this is proved for several observables in free space. The absence of direct interatomic interactions often makes calculations simpler in the multipolar representation.

### 4.4.3 In need of a local-field model

The quantum multipolar Hamiltonian will have an atomic and a radiative part, as well as two interaction terms:

$$H_{\text{multi}} = H_{\text{rad}} + \sum_m \left[ H_{\text{at}}^{(m)} + V_{\text{P}}^{(m)} + V_{\text{M}}^{(m)} \right]. \quad (4.55)$$

The aim of this section is to discuss the form of the atomic Hamiltonian  $H_{\text{at}}^{(m)}$ ; in section 4.4.4 the Hamiltonian will be given in second-quantization notation.

The Hamiltonian for a guest atom inside the dielectric is

$$H_{\text{at}}^{(m)} = \frac{1}{2m_e} \sum_j p_{mj}^2 + \int d\mathbf{r} P_{gm}^2 / [2\varepsilon_0\varepsilon(\mathbf{r})]. \quad (4.56)$$

This Hamiltonian differs from free-space atomic Hamiltonian. This point is missed if one introduces a “two-level atom” with known transition frequency into the dielectric. The second term in (4.56) is the potential energy and it consists of longitudinal (Coulomb) and transverse interactions between the charges. The potential energy is reduced by a factor  $\varepsilon(\mathbf{r})$  as compared to free space. This reduction factor is well known for dielectric-filled capacitors.

Until now it was tacitly assumed that  $\varepsilon(\mathbf{r})$  is a given function that characterizes the dielectric in the absence of the guest atoms. Still, it could be that  $\varepsilon(\mathbf{r})$  is different locally from the situation without the guest atoms. Suppose that one neglects local changes on the grounds that the dielectric function is a macroscopically averaged quantity that does not change on atomic length scales. Then  $\varepsilon(\mathbf{r})$  must be unchanged by introducing a guest atom and the Hamiltonian can be approximated by

$$H_{\text{at}}^{(m)} \approx \frac{1}{2m_e} \sum_j p_{mj}^2 + \frac{1}{2\varepsilon_0\varepsilon(\mathbf{R}_m)} \int d\mathbf{r} P_{gm}^2. \quad (4.57)$$

If the guest atom were a hydrogen atom, then its Bohr radius and dipole moments would increase by a factor  $\varepsilon(\mathbf{R}_m)$  and its energy levels would be reduced by the same factor, according to this Hamiltonian. Any visible line in free space would then be shifted to the infrared in a dielectric. The dielectric would have a huge effect on the atom's electronic properties.

Indeed, it has been known for a long time that wave functions of impurity atoms can be severely modified by the medium from their values in free space [89]. This has important consequences, for example when doping solid silicon with phosphorus to make an  $n$ -type semiconductor. The high dielectric constant of Si ( $\varepsilon = 11.7$ ) reduces the potential energy between the outermost electron and the rest of the P atom, so that the electron can enter the conduction band relatively easily, leaving a  $P^+$ -ion [90]. Such changes in atomic wave functions and energies with a broad term will be called electronic effects of the dielectric on the atom.

Atomic lifetime changes and line shifts can have electronic and photonic explanations. Changes due to modified atomic dipole moments are electronic effects. A macroscopic description of the dielectric is too coarse-grained to say anything quantitative about electronic effects. It is the photonic effects, the changes due to altered properties of the electromagnetic field, which are of primary interest here. However, only if the electronic changes of the atoms are somehow either absent or accounted for, can one study the photonic effects. For example, in a recent study [91] of local-field effects [4, 11, 92–95] in spontaneous emission, it was important that atomic spectra (and therefore dipole moments) do not change while varying the refractive index. As a second example, the recently observed fivefold reduction of spontaneous-emission rates inside photonic crystals [35] is a photonic effect, since possible changes in dipole moments were divided out by choosing a reference sample with identical electronic effects [96]. Some earlier observations of long lifetimes in photonic crystals must be attributed to electronic effects, according to recent calculations [97].

In practice, line shifts are often much smaller than in the estimate in Eq. (4.57) where local modifications of the dielectric function are neglected. As a first example of local modifications, outer electronic states of the atom are more affected by the dielectric than the core electrons. In other words, outer electrons feel a different dielectric function than the inner electrons. The latter are screened by the former. As a second example, in some experiments the guest atoms are surrounded by a low-index molecular complex to separate electronic from photonic effects of the dielectric [91].

The absence of large electronic effects of the dielectric on the atomic properties can not be explained within the present macroscopic quantization theory. Only from microscopic theories of the dielectric can one derive that the dielectric can not be where the guest atoms are. In the present context of macroscopic quantization, a model is needed that does justice to the above two examples of local modifications of the dielectric by the guest atoms. A real-cavity model is assumed, with atomic-sized holes in the dielectric function at the atomic positions  $\mathbf{R}_m$ . In this model, the atomic Hamiltonian (4.56) can be approximated by (4.57) with  $\varepsilon(\mathbf{R}_m)$  equal to 1. The model captures the observed absence of large electronic effects of the dielectric on atomic properties of interest, but at the same time the model has consequences for photonic properties: the local changes in  $\varepsilon$  will

give local changes in the mode functions  $f_\lambda$ , and therefore in the dipole coupling (4.62) and also in the spontaneous-emission rates of the guest atoms [11]. Some quantitative consequences of adopting the real-cavity model will be given in chapter 5.

This chapter first presented the dielectric without guest atoms. Later the guest atoms were introduced. In this section we see that actually there must be three steps: first calculate the mode functions of the inhomogeneous dielectric. Second, make atomic-sized holes at the positions where the guest atoms will be introduced. Determine the locally modified mode functions of the dielectric with the tiny cavities. The third and last step is the introduction of the guest atoms themselves, which couple to the electromagnetic field through the modes of step two.

#### 4.4.4 Quantum multipolar interaction Hamiltonian

The goal is now to rewrite important terms of the Hamiltonian (4.53) into a second-quantization description, where macroscopic quantization has been applied to the electromagnetic field and microscopic quantization to the guest atoms.

First start with the atomic Hamiltonian (4.57) where  $\varepsilon(\mathbf{R}_m)$  is put to 1. Following (standard) quantum mechanics, the electron coordinates  $\mathbf{r}_{mj}(t)$  and their canonical momenta  $\mathbf{p}_{mj}(t)$  (4.51), as well as the polarization and magnetization fields  $\mathbf{P}_g$  and  $\mathbf{M}_g$ , become time-independent operators that work on the time-dependent atomic wave functions. The single-atom wave functions can be expanded in terms of eigenfunctions (labelled  $k$ ) of the atomic Hamiltonian:

$$\Psi_m(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_{Z_m}; t) = \sum_k c_{mk}(t) \psi_{mk}(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_{Z_m}). \quad (4.58)$$

The time-dependence is in the probability amplitudes  $c_{mk}(t)$ . The time-independent functions  $\psi_{mk}$  are eigenfunctions of the atomic Hamiltonian (4.57) with  $\varepsilon(\mathbf{R}_m) = 1$ .

Second-quantization notation can now be introduced by promoting the probability amplitudes  $c_{mk}(t)$  and  $c_{mk}^*(t)$  in (4.58) to become annihilation and creation operators with standard anti-commutation relations. These operators become the atomic canonical variables in the second-quantization picture. (One could even go back and start with a Lagrangian that identifies  $\Psi_m$  and  $\Psi_m^*$  as canonical conjugates [98].) The atomic operators can be written as sums over matrix elements. For example, the atomic Hamiltonian of atom  $m$  in standard second-quantization notation is  $H_{\text{at}}^{(m)} = \sum_k E_k^{(m)} c_{mk}^\dagger c_{mk}$ . Within the real-cavity model that was adopted, all atomic energy levels  $E_{mk}$  in this equation are numerically the same as in free space. This completes the quantization and second quantization of the atomic Hamiltonian.

Three terms of the quantum multipolar interaction Hamiltonian (4.55) must still be given in second-quantization notation. The electromagnetic field can again be quantized by expanding the vector potential and its canonically conjugate field into a complete set of true modes. The subsequent quantization steps are as presented in section 4.2.2 in the absence of guest atoms. The radiative part of the Hamiltonian becomes  $H_{\text{rad}} = \sum_\lambda \hbar \omega_\lambda (a_\lambda^\dagger a_\lambda + \frac{1}{2})$ . The form of this Hamiltonian has not changed by the presence of guest atoms, but there is a slight shift in its interpretation, since the polarization of the guest atoms is included in the conjugate field.

The interaction  $V_P^{(m)}$  is associated with the polarization field of the guest atom  $m$  and has the form

$$\begin{aligned} V_P^{(m)} &= - \int d\mathbf{r} \frac{\mathbf{P}_{gm}(\mathbf{r}) \cdot \mathbf{D}(\mathbf{r})}{\varepsilon_0 \varepsilon(\mathbf{r})} \\ &= -i \sum_{\lambda} \sum_{k,k'} \sqrt{\frac{\hbar \omega_{\lambda}}{2\varepsilon_0}} \left[ a_{\lambda} c_{mk}^{\dagger} c_{mk'} \int d\mathbf{r} \mathbf{P}_{gm,kk'}(\mathbf{r}) \cdot \mathbf{f}_{\lambda}(\mathbf{r}) - \text{H.c.} \right], \end{aligned} \quad (4.59)$$

where “H.c.” stands for the Hermitian conjugate. The quantity  $\mathbf{P}_{gm,kk'}(\mathbf{r})$  is the matrix element of the polarization field  $\mathbf{P}_{gm}(\mathbf{r})$  with respect to states  $\psi_{mk}$  and  $\psi_{mk'}$  of atom  $m$ . These atomic states are unaffected by the dielectric, at least in the empty-cavity model that was adopted in section 4.4.3 and which is also assumed here. Thus, the polarization field and its matrix elements are functions of the atomic variables alone, whereas the displacement field depends solely on the electromagnetic canonical variables. The two interacting fields are therefore canonically independent. The interpretation of the interaction is more subtle, since in the definition (4.46) of the displacement field the polarization of the guest atoms is included. The interaction  $V_P^{(m)}$  therefore includes a self-interaction of the polarization field.

The second and last interaction term  $V_M^{(m)}$  in (4.55) is associated with the reduced magnetization field of the  $m^{\text{th}}$  guest atom:

$$\begin{aligned} V_M^{(m)} &= - \int d\mathbf{r} \mathbf{M}'_{gm}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}) \\ &= - \sum_{\lambda} \sum_{k,k'} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega_{\lambda}}} \left\{ a_{\lambda} c_{mk}^{\dagger} c_{mk'} \int d\mathbf{r} \mathbf{M}'_{gm,kk'}(\mathbf{r}) \cdot [\nabla \times \mathbf{f}_{\lambda}(\mathbf{r})] + \text{H.c.} \right\}. \end{aligned} \quad (4.60)$$

Again, the empty-cavity model entails that matrix elements  $\mathbf{M}'_{gm,kk'}$  of the reduced magnetization field do not depend on the dielectric. The magnetic field and the reduced magnetization field are canonically independent and  $\mathbf{M}'_g$  is not included in the definition of  $\mathbf{B}$ . All four terms in the quantum multipolar Hamiltonian (4.55) have now been given in second-quantization notation.

#### 4.4.5 Dipole approximation

The guest atoms are much smaller than an optical wavelength. One cannot spatially probe the structure of the atom with light. If light can not tell the difference, one can make the well-known assumption that the polarization and magnetization fields associated with the atom are concentrated in its center of mass  $\mathbf{R}_m$  (the nucleus, say). Mathematically, this means that the integrands in Eq. (4.44) are approximated by their values in  $u = 0$ . These two values are the first terms of two infinite Taylor expansions in terms of the variable  $u$ . The dipole approximation is made by keeping only the first term. The next terms would describe the quadrupole interaction. In the dipole approximation, the magnetization

(reduced or not) becomes identically zero and the polarization field becomes

$$\begin{aligned} \mathbf{P}_{gm}(\mathbf{r}) &= -e \delta(\mathbf{r} - \mathbf{R}_m) \sum_{k,k'} c_{mk}^\dagger \langle \psi_{mk} | \sum_j (\mathbf{r}_{mj} - \mathbf{R}_m) | \psi_{mk'} \rangle c_{mk'} \\ &\equiv \delta(\mathbf{r} - \mathbf{R}_m) \sum_{k,k'} c_{mk}^\dagger \boldsymbol{\mu}_{kk'}^{(m)} c_{mk'}. \end{aligned} \quad (4.61)$$

The last equality defines the atomic dipole matrix elements  $\boldsymbol{\mu}_{kk'}^{(m)}$  of the guest atom  $m$ . These matrix elements are independent of the dielectric environment in the real-cavity model. With equation (4.59), it follows that in the dipole approximation the interaction energy of an atom with the electromagnetic field in an inhomogeneous dielectric equals

$$\begin{aligned} V_{\text{dip}}^{(m)} &= - \sum_{kk'} c_{mk}^\dagger c_{mk'} \boldsymbol{\mu}_{kk'}^{(m)} \cdot \mathbf{D}(\mathbf{R}_m) / \varepsilon_0 \\ &= -i \sum_{\lambda} \sum_{kk'} \sqrt{\frac{\hbar \omega_{\lambda}}{2 \varepsilon_0}} \left[ a_{\lambda} c_{mk}^\dagger c_{mk'} \boldsymbol{\mu}_{kk'}^{(m)} \cdot \mathbf{f}_{\lambda}(\mathbf{R}_m) - \text{H.c.} \right]. \end{aligned} \quad (4.62)$$

In the first identity it was used that  $\varepsilon(\mathbf{R}_m)$  equals unity in the real-cavity model (adopted in section 4.4.3). This gives the important result that inside an inhomogeneous dielectric, a dipole couples to the field  $-\mathbf{D}/\varepsilon_0$ , but *necessarily at a position where the dielectric function equals one*. This is a generalisation of the free-space dipole-coupling [84, 98, 99] and a necessary qualification of the dipole coupling found in [13, 14].

The dipole approximation is excellent for real atoms. For the guest atoms one can also think of other (larger) quantum systems in interaction with the electromagnetic field. “Artificial atoms”, also known as quantum dots, are an example. Quantum dots are semiconductor nanostructures tens of nanometers in size or less in all three dimensions. Quantum dots are much larger than real atoms and so their dipole moments can be much larger as well (see for example [100] and references therein). Because of their larger size, quadrupole moments are more important for quantum dots than for real atoms, especially when excited in their near field by a scanning near field optical microscope [101].

## 4.5 Inhomogeneous magnetic media

In the past few years, old and nearly forgotten ideas about so-called left-handed materials [102] have become the subject of intense scientific discussions after a prediction that a perfect lens could be made with them [103]. Left-handed materials do not exist in nature and have the exotic property that both their relative dielectric functions  $\varepsilon(\mathbf{r})$  and their relative magnetic permeabilities  $\mu(\mathbf{r})$  are negative. Their index of refraction is also negative and this leads to many peculiar properties. They will also influence spontaneous-emission rates of nearby guest atoms in different ways than their right-handed counterparts, according to a recent paper [104]. Without diving into this subject here, left-handed materials make it interesting to generalize the formalism of this chapter to dielectrics with inhomogeneous magnetic properties as well. Such generalizations were already considered in [13, 14] and are relatively straightforward, for reasons discussed below.

When  $\mu$  becomes position-dependent, then the only term that will change in the classical multipolar Hamiltonian (4.53) is the magnetic field energy  $\int d\mathbf{r} \frac{\mathbf{B}^2(\mathbf{r})}{2\mu_0\mu(\mathbf{r})}$ . (Confusion should not arise about the symbol  $\mu$  that is used as the magnetic permeability and as the magnitude of a dipole  $\boldsymbol{\mu}$ .) The quantum mechanical description can again be carried out by choosing the generalized Coulomb gauge. Without guest atoms, the vector potential satisfies the source-free wave equation

$$\nabla \times \left[ \frac{1}{\mu(\mathbf{r})} \nabla \times \mathbf{A}(\mathbf{r}) \right] + \frac{\varepsilon(\mathbf{r})}{c^2} \ddot{\mathbf{A}}(\mathbf{r}) = 0. \quad (4.63)$$

The electromagnetic field can be expanded in terms of new modes different from the modes  $\mathbf{f}_\lambda$ . The new normal modes  $\mathbf{m}_\lambda$  are the harmonic solutions of the wave equation (4.63). These new modes are also generalized transverse, just like the modes  $\mathbf{f}_\lambda$  of the nonmagnetic medium. This is what makes the generalization of the formalism to magnetic media relatively simple. In second quantization notation, the electromagnetic field energy becomes  $\sum_\lambda \hbar\omega_\lambda \left( d_\lambda^\dagger d_\lambda + \frac{1}{2} \right)$ , where  $d_\lambda^\dagger$  is the creation operator of a photon in the mode  $\mathbf{m}_\lambda(\mathbf{r})$ . The new modes can have mode profiles that differ much from any of the modes  $\mathbf{f}_\lambda(\mathbf{r})$ , but otherwise the theory of the medium is not much different. In the electric and magnetic interactions (4.59) and (4.60) of the electromagnetic field with guest atoms, the modes  $\mathbf{f}_\lambda$  can just be replaced by the  $\mathbf{m}_\lambda$  and the operators  $c_\lambda^{(\dagger)}$  by  $d_\lambda^{(\dagger)}$  in order to take both the electric and magnetic properties of the medium into account. As for nonmagnetic media, in the dipole approximation the magnetic interaction (4.60) is zero; the electric dipole interaction (4.59) dominates, except for optical transitions with zero dipole moments. When dipole moments are nonzero, the main effect of the dielectric becoming magnetic comes from the change in the mode functions. When in the future left-handed materials will be made with negligible dispersion and absorption in the frequency interval where  $\varepsilon$  and  $\mu$  are negative, then the present formalism will also be applicable to those exotic materials.

## 4.6 Dipole-coupling controversy

Many papers appeared in the nineteen-eighties about the equivalence of the minimal-coupling and the multipolar Hamiltonian in free space, for example [98, 99, 105–114]. The Hamiltonians sometimes lead to different results in calculations. Some authors argued that the minimal-coupling Hamiltonian was to be preferred [106], while others proposed to refrain from using gauge-dependent equations to stop the confusion [109]. In the multipolar picture, a controversy arose whether a dipole in free space couples to minus the displacement field ( $-\boldsymbol{\mu} \cdot \mathbf{D}/\varepsilon_0$ , see [99]) or to the transverse part of the electric field ( $-\boldsymbol{\mu} \cdot \mathbf{E}^T$ , see [115]). The book by Cohen-Tannoudji *et al.* helped a lot to settle the dust [84]. It is probably useful to briefly mention a few reasons why confusion is so likely to arise in a situation which in principle is not difficult.

In the first place, it is important to distinguish canonical variable transformations (new variables, same Hamiltonian and states) from unitary changes of picture (new Hamiltonian, new states, same expectation values). This distinction was excellently presented in [99].

Unlike canonical transformations, picture changes have no classical analogues. Confusion is likely to arise when after a canonical change a Hamiltonian has exactly the same form as after a picture change [99]. This is one reason why a controversy about the dipole coupling term could arise.

A second source of confusion is related to approximations. It was found in section 4.4.5 that a dipole at position  $\mathbf{R}$  inside a dielectric couples to the displacement field

$$-\mathbf{D}(\mathbf{R})/\varepsilon_0 = -[\mathbf{E}(\mathbf{R}) + \mathbf{P}_g(\mathbf{R})]/\varepsilon_0, \quad (4.64)$$

where  $\mathbf{P}_g$  is the polarization field of the guest atom itself; the equality in (4.64) holds because the empty-cavity model was assumed so that  $\varepsilon(\mathbf{R})$  equals one. In free space, a dipole couples to the same field (4.64). Why then are there still authors that state that in free space [115] or in a dielectric [11] a dipole couples to minus the electric field? For the dielectric with guest atoms, the displacement field operator can be expanded as

$$\mathbf{D}(\mathbf{r}) = i\varepsilon_0\varepsilon(\mathbf{r}) \sum_{\lambda} \sqrt{\frac{\hbar\omega_{\lambda}}{2\varepsilon_0}} \left[ a_{\lambda} \mathbf{f}_{\lambda}(\mathbf{r}) - a_{\lambda}^{\dagger} \mathbf{f}_{\lambda}^*(\mathbf{r}) \right]. \quad (4.65)$$

The form of this operator is the same as in a dielectric without guest atoms [minus the equation (4.25b)], except that the equations of motion of the creation- and annihilation operators in (4.65) have terms involving the atomic variables, which the equations of motion of their counterparts  $a_{\lambda}^{(0)}$  and  $a_{\lambda}^{(0)\dagger}$  do not have. If one approximates the displacement field (4.65) and its time-derivatives by assuming free-space time-dependence, then for positions where  $\varepsilon$  equals one, the displacement field (4.65) is equal to the electric field in the absence of the guest atoms. In other words, if the guest atoms are taken into account in Maxwell's equations, then one finds a dipole coupling to the displacement field, whereas a coupling to the electric field is found when guest atoms are left out of Maxwell's equations. It depends on the observable under study whether the difference between the two dipole couplings can be neglected or not. An example where only one dipole coupling is correct will be given in the next chapter in section 5.2.3.

## 4.7 Summary and discussion

The aim of the chapter was to find a Hamiltonian of guest atoms in an inhomogeneous dielectric, with a multipolar interaction between the atoms and the field. The multipolar Hamiltonian is simpler than the minimal-coupling Hamiltonian, because in the former all interactions between the atoms are mediated by the retarded electromagnetic field. The main results of this chapter are therefore the quantum multipolar interaction Hamiltonian (4.55) together with its dipole approximation (4.62). With this Hamiltonian, one can study how a dielectric environment can change quantum optical processes of resonant atoms.

Another important result is that the macroscopic description of the dielectric could only be tied up to the microscopic description of the atoms by assuming that the dielectric function has the value 1 at the atomic positions. If not, the atoms in the dielectric would have huge frequency shifts. The local modification of the dielectric function will also change the dipole coupling, because mode functions change locally (see also chapter 5). If

one would start with the same two- or three-level description of the guest atoms as in free space, one implicitly already assumes such a model for the dielectric function. Consistency requires to also choose a local-field model when calculating the mode functions in the dipole interaction (4.62).

In order to obtain the central results of this chapter, it was necessary to first quantize the electromagnetic field in the dielectric without guest atoms. This has certainly been carried out before, but after explaining why real optical mode functions can be used whenever  $\varepsilon(\mathbf{r})$  is real, the quantization becomes simpler than found in [11, 13, 86]. In particular, only when real mode functions are chosen are the associated generalized positions and momenta Hermitian. The field that must be so quantized is the field of the dielectric with empty cavities at the positions where the guest atoms are to be placed.

The guest atoms were introduced into the theory such that Maxwell's equations hold with the atomic charge and current densities as source terms. A minimal-coupling Lagrangian that gives rise to these equations was easily written down. However, after choosing a generalized Coulomb gauge, it was not directly clear how to obtain all Maxwell's equations in this particular gauge. In the important appendix B, the mathematical question was studied what it means to take a functional derivative after choosing a gauge. It was shown that the derivative of the minimal-coupling Lagrangian could be found by solving a certain electrostatical potential problem. As a consequence, the gauge-independent Maxwell's equations were found indeed to hold in the generalized Coulomb gauge as well.

The application of the Power-Zienau-Woolley transformation to the minimal-coupling Lagrangian was shown to produce the multipolar Lagrangian, after the generalized Coulomb gauge had been chosen. Actually, the gauge was chosen earlier than strictly necessary: the choice could have been postponed until the canonical momenta were determined from the multipolar Lagrangian. The story would have been simpler up to that point. The difficulty to find all Maxwell's equations would then show up only after obtaining the multipolar Lagrangian. The reason to first choose the gauge and then do the PZW transformation, is that it more clearly shows that the difficulty to find all Maxwell's equations was a consequence of choosing the gauge, rather than a consequence of the PZW transformation.

More generally, the presentation given in this chapter is one among the many possibilities. When going from a classical minimal-coupling Lagrangian to a quantum mechanical multipolar Hamiltonian, one has to make four steps: one step is to choose a gauge, another step is to transform the theory to the multipolar formalism. Yet another step is made when going from a Lagrangian to a Hamiltonian; quantization and second quantization together are step number four. These are now given in the order in which they occurred in this chapter, but the steps can be interchanged. Not all of the 24 permutations are convenient. All routes should lead to equivalent final results. In particular, it was shown in detail in [111] that step two and three can be interchanged for free space: the PZW transformation of the minimal-coupling Lagrangian is equivalent to a picture change of the minimal-coupling Hamiltonian. The equivalence will also hold for inhomogeneous dielectrics.

## Chapter 5

# Point scatterers and quantum optics in inhomogeneous dielectrics

*A point-scattering formalism is set up which is very convenient when studying quantum optical processes in dielectrics. The starting point is the multipolar Hamiltonian for guest atoms inside inhomogeneous dielectrics that was derived in chapter 4. The formalism will first be used to calculate position-dependent single-atom spontaneous-emission rates. Then it will be shown how two-atom superradiance can be strongly modified inside an inhomogeneous dielectric. The emphasis in the chapter will be on the influence that the dielectric has on the processes which are well-known in free space.*

### 5.1 Introduction

The spontaneous-emission rate of an atom depends on its dielectric environment [1] and in particular on the precise position of the atom if the medium is inhomogeneous [11, 41, 116–118]. The spontaneous-emission rate is proportional to the local optical density of states of the medium [53]. Local densities of states were determined from the Green function in chapters 2 and 3. One of the main objectives in those chapters was to find the Green functions for inhomogeneous media with the use of multiple-scattering theory. Here, the goal will be to describe optical processes such as spontaneous emission and superradiance in terms of the Green functions, rather than to calculate these Green functions themselves.

The system under study in this chapter is the same as in the previous chapter 4: a collection of resonant guest atoms in a non-absorbing inhomogeneous dielectric. Based on the final results of that chapter, it will be studied how the guest atoms interact with the medium and with each other. The concept of a point scatterer proved very fruitful in the study of multiple-scattering of classical light in free space [68, 69, 94]. Here, the point-scattering formalism will be put to use in quantum optics of inhomogeneous dielectrics.

Spontaneous-emission rates and frequency shifts of individual atoms are derived, with emphasis on the spatial properties of light scattering and emission.

Not only single-atom properties are changed. The dielectric environment also modifies cooperative effects of nearby atoms. The canonical example of cooperative effects is two-atom superradiance. Here, the effect of an inhomogeneous medium on two-atom superradiance will be studied. The precise measurement of two-atom superradiance in free space as a function of distance is a fundamental test for quantum electrodynamics [119]. The effects calculated here are a test for macroscopic quantization theories for inhomogeneous dielectrics described by a nondispersive and real dielectric function  $\varepsilon(\mathbf{r})$ . A closely related subject of fundamental and also of technological interest is the influence of the dielectric environment on the transfer of an optical excitation from a donor to an acceptor atom. It has been studied theoretically how two-atom interactions are influenced by an optical microcavity [120–122]. Here the resonant modes of the cavity play a dominant role, the modes with eigenfrequencies close or equal to the atomic transition frequency. Experiments have shown that the dipole-dipole interaction is increased when the atoms are placed in a cavity at positions  $\mathbf{R}_1$  and  $\mathbf{R}_2$  where resonant optical modes have their maxima [123].

The chapter has the following structure: in section 5.2 the point-scattering model for interacting guest atoms is introduced; section 5.3 discusses medium-induced modifications of single-atom properties such as spontaneous-emission rates and elastic scattering. Generalization to a finite number of host atoms is discussed in section 5.4. The formalism is applied to two-atom superradiance in section 5.5. As a special case, superradiance in a homogeneous dielectric is discussed in section 5.6. This is followed by a qualitative discussion in section 5.7 on line shifts and dipole-dipole interactions in inhomogeneous dielectrics and in particular in photonic crystals. Conclusions are drawn in section 5.8.

## 5.2 Atoms as point sources and as point scatterers

The light intensity operator is important in the analysis of almost all quantum optical experiments. In terms of the electric field it has the form [15]

$$I(\mathbf{r}, t) = 2\varepsilon_0 c \mathbf{E}^{(-)}(\mathbf{r}, t) \cdot \mathbf{E}^{(+)}(\mathbf{r}, t), \quad (5.1)$$

where  $\mathbf{E}^{(+)}$  is the positive-frequency part of the electric field operator, and  $\mathbf{E}^{(-)}$  is its Hermitian conjugate. The intensity operator is also important now that light scattering and emission by guest atoms in dielectrics is studied: measurable intensities correspond to expectations values of the intensity operator with respect to the initial state of both the electromagnetic field and of the guest atoms. The goal in this section is to find a Lippmann-Schwinger equation for the electric field from which its time and position dependence can be derived. The result can directly be inserted in equation (5.1) so that intensities (and intensity correlations) can be found. A Hamiltonian will be proposed in section 5.2.1; equations of motion are solved in sections 5.2.2 and 5.3.1.

### 5.2.1 The Hamiltonian

The Hamiltonian for  $N$  atoms inside an inhomogeneous dielectric is the sum of a field part, an atomic part, and an interaction part between field and atoms. A multipolar interaction is assumed and the dipole approximation (4.62) is made. Since in the coming derivations the field operator  $\mathbf{D}(\mathbf{r}, t)/[\varepsilon_0 \varepsilon(\mathbf{r})]$  plays an important role, it will be abbreviated in the following as

$$\mathbf{F}(\mathbf{r}, t) \equiv \mathbf{D}(\mathbf{r}, t)/[\varepsilon_0 \varepsilon(\mathbf{r})]. \quad (5.2)$$

The field operator  $\mathbf{F}$  will be referred to as the “Field”. At the positions  $\mathbf{R}_m$  of the guest atoms, the Field equals the displacement field as defined in equation (4.46), because  $\varepsilon(\mathbf{r}) = 1$  at those spots. On the other hand, away from the atoms, in a detector for example, the Field equals the electric field. The details of this subtle distinction can be found in chapter 4 and become important in section 5.2.3. The Field is the sum of a positive-frequency part  $\mathbf{F}^{(+)}$  containing only annihilation operators and its Hermitian conjugate  $\mathbf{F}^{(-)}$ ; it has a simple expansion in terms of the normal mode solutions of the electric field:

$$\mathbf{F}(\mathbf{r}, t) = \mathbf{F}^{(+)}(\mathbf{r}, t) + \mathbf{F}^{(-)}(\mathbf{r}, t) = i \sum_{\lambda} \sqrt{\frac{\hbar \omega_{\lambda}}{2 \varepsilon_0}} \left[ a_{\lambda}(t) \mathbf{f}_{\lambda}(\mathbf{r}) - a_{\lambda}^{\dagger}(t) \mathbf{f}_{\lambda}^*(\mathbf{r}) \right]. \quad (5.3)$$

In the absence of the atoms, the time dependence of the creation- and annihilation operators in (5.3) would be harmonic and the Field would be equal to the electric field  $\mathbf{E}^{(0)}$ . But now the atoms are there and the total Hamiltonian is

$$\mathcal{H} = \mathcal{H}_F + \mathcal{H}_A + \mathcal{H}_{AF}, \quad \text{with} \quad (5.4a)$$

$$\mathcal{H}_F = \sum_{\lambda} \hbar \omega_{\lambda} a_{\lambda}^{\dagger} a_{\lambda} \quad (5.4b)$$

$$\mathcal{H}_A = \sum_{m=1}^N \hbar \Omega_m b_m^{\dagger} b_m \quad (5.4c)$$

$$\mathcal{H}_{AF} = - \sum_{m=1}^N \boldsymbol{\mu}_m \cdot \mathbf{F}(\mathbf{R}_m) = \sum_{m, \lambda} (b_m + b_m^{\dagger})(g_{\lambda m} a_{\lambda} + g_{\lambda m}^* a_{\lambda}^{\dagger}). \quad (5.4d)$$

The atomic frequencies  $\Omega_m$  and transition dipole moments  $\boldsymbol{\mu}_m$  may be all different, either because the guest atoms are of different species or because identical atoms feel different electronic modifications due to the dielectric. Only when an empty-cavity model applies can electronic modifications be neglected, recall section 4.4.3. The frequencies  $\Omega_m$  are assumed real, which means that nonradiative broadening is neglected. The atoms are very simply described as harmonic oscillators with frequencies  $\Omega_m$ . This is a good approximation as long as saturation effects of the upper atomic state can be neglected. (For a more realistic modelling of the internal states of atoms one could turn to [124].) The atomic dipole moments  $\boldsymbol{\mu}_m$  are assumed to have a fixed orientation. This assumption is better for molecules or quantum dots in a solid surrounding than for atoms in the gas phase. For convenience, the name “atoms” will be used for the guest in the dielectric. The operators  $b_m^{\dagger}$  create atomic excitations by annihilating an atom in the ground state while at the same

time creating the atom in the excited state: in terms of the operators of section 4.4.5,  $b_m^\dagger$  equals  $c_{m1}^\dagger c_{m0}$ . The excitation ground state  $|0\rangle$  for the operators  $b_m^\dagger$  corresponds to the electronic ground state  $c_{m0}^\dagger|0\rangle$ .

The coupling constants between atom  $m$  and optical mode  $\lambda$  in Eq. (5.4d) are defined as

$$g_{\lambda m} = -i\sqrt{\frac{\hbar\omega_\lambda}{2\varepsilon_0}}\boldsymbol{\mu}_m \cdot \mathbf{f}_\lambda(\mathbf{R}_m). \quad (5.5)$$

The field part of the Hamiltonian is a sum (or integral) over harmonic oscillators corresponding to the harmonic solutions (“true modes”) of the Maxwell equations for the inhomogeneous dielectric in the absence of the atoms.

### 5.2.2 Integrating out atomic dynamics

The goal of this section is to derive a Lippmann-Schwinger equation for the Field inside the inhomogeneous dielectric in the presence of the  $N$  guest atoms. Heisenberg’s equation of motion leads to the following equations of motion for the Field operators:

$$\dot{a}_\lambda(t) = -i\omega_\lambda a_\lambda(t) - (i/\hbar) \sum_m g_{\lambda m}^* [b_m(t) + b_m^\dagger(t)] \quad (5.6a)$$

$$\dot{a}_\lambda^\dagger(t) = i\omega_\lambda a_\lambda^\dagger(t) + (i/\hbar) \sum_m g_{\lambda m} [b_m(t) + b_m^\dagger(t)]. \quad (5.6b)$$

The Field operators are coupled to the atomic operators and the operators of atom  $m$  satisfy the equations

$$\dot{b}_m(t) = -i\Omega_m b_m(t) - (i/\hbar) \sum_\lambda [g_{\lambda m} a_\lambda(t) + g_{\lambda m}^* a_{\lambda m}^\dagger(t)] \quad (5.7a)$$

$$\dot{b}_m^\dagger(t) = i\Omega_m b_m^\dagger(t) + (i/\hbar) \sum_\lambda [g_{\lambda m} a_\lambda(t) + g_{\lambda m}^* a_{\lambda m}^\dagger(t)]. \quad (5.7b)$$

Now take the Laplace transform (or positive-time Fourier transform) of the equations of motion (with argument  $-i\omega$ ), for example

$$b_m(\omega) \equiv \int_0^\infty dt e^{i\omega t} b_m(t). \quad (5.8)$$

Here and in the following the frequency  $\omega$  is assumed to contain an infinitesimally small positive imaginary part so that the above integral is well-defined. The equations are algebraic after the transformation.

One can assume that at time zero, the annihilation operators  $a_\lambda$  coincide with the  $a_\lambda^{(0)}$ , the operators in the absence of the guest atoms. The  $a_\lambda^{(0)}$  have simple harmonic equations of motion  $\dot{a}_\lambda^{(0)} + i\omega_\lambda a_\lambda^{(0)} = 0$ . After partially integrating, the Laplace transform of this equation becomes

$$-i(\omega - \omega_\lambda) a_\lambda^{(0)}(\omega) = a_\lambda^{(0)}(t=0). \quad (5.9)$$

Also in Laplace language, the equations for the atomic operators become

$$b_m(\omega) = \frac{ib_m(0)}{\omega - \Omega_m} + \frac{\hbar^{-1}}{\omega - \Omega_m} \sum_{\lambda} \left[ g_{\lambda m} a_{\lambda}(\omega) + g_{\lambda m}^* a_{\lambda m}^{\dagger}(\omega) \right] \quad (5.10a)$$

$$b_m^{\dagger}(\omega) = \frac{ib_m^{\dagger}(0)}{\omega + \Omega_m} - \frac{\hbar^{-1}}{\omega + \Omega_m} \sum_{\lambda} \left[ g_{\lambda m} a_{\lambda}(\omega) + g_{\lambda m}^* a_{\lambda m}^{\dagger}(\omega) \right]. \quad (5.10b)$$

The right-hand sides of Eqs. (5.10a) and (5.10b) are used to replace  $b_m(\omega)$  and  $b_m^{\dagger}(\omega)$  in the Laplace transforms of the equations (5.6a) and (5.6b) for the Field operators. In doing this, the atomic dynamics is integrated out. With the use of the identity (5.9), one obtains for the annihilation operators of the Field

$$\begin{aligned} a_{\lambda}(\omega) &= a_{\lambda}^{(0)}(\omega) + \frac{i\hbar^{-1}}{\omega - \omega_{\lambda}} \sum_m g_{\lambda m}^* \left[ \frac{b_m(0)}{\omega - \Omega_m} + \frac{b_m^{\dagger}(0)}{\omega + \Omega_m} \right] \\ &+ \frac{\hbar^{-2}}{\omega - \omega_{\lambda}} \sum_{m, \lambda'} g_{\lambda m}^* \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \left[ g_{\lambda' m} a_{\lambda' m}(\omega) + g_{\lambda' m}^* a_{\lambda' m}^{\dagger}(\omega) \right]. \end{aligned} \quad (5.11a)$$

For the creation operators, a similar equation is found, be it with some essential sign changes:

$$\begin{aligned} a_{\lambda}^{\dagger}(\omega) &= a_{\lambda}^{(0)\dagger}(\omega) - \frac{i\hbar^{-1}}{\omega + \omega_{\lambda}} \sum_m g_{\lambda m} \left[ \frac{b_m(0)}{\omega - \Omega_m} + \frac{b_m^{\dagger}(0)}{\omega + \Omega_m} \right] \\ &- \frac{\hbar^{-2}}{\omega + \omega_{\lambda}} \sum_{m, \lambda'} g_{\lambda m} \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \left[ g_{\lambda' m} a_{\lambda' m}(\omega) + g_{\lambda' m}^* a_{\lambda' m}^{\dagger}(\omega) \right]. \end{aligned} \quad (5.11b)$$

Evidently, the optical modes are no longer independent because of the interaction with the atoms. The three terms in the right-hand sides of Eqs. (5.11a) and (5.11b) can respectively be identified with three reasons why there can be light in mode  $\lambda$ : firstly, because there is light in the undisturbed mode that has not “seen” the atom; secondly, because the atom can emit light into the mode  $\lambda$ ; the third term describes transitions of light in and out of the mode  $\lambda$  to and from modes  $\lambda'$ , due to scattering off one of the guest atoms. Since the relations (5.11a) and (5.11b) are implicit rather than explicit solutions for the operators, the identification of terms in the equations with scattering and emission processes can only be approximate.

These results for the creation and annihilation operators can be directly used with Eq. (5.3) to find the following equation for the Field (including both positive- and negative-frequency parts)

$$\begin{aligned} \mathbf{F}(\mathbf{r}, \omega) &= \mathbf{E}^{(0)}(\mathbf{r}, \omega) \\ &+ \sum_m \mathbf{K}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{S}_m(\omega) + \sum_m \mathbf{K}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{V}_m(\omega) \cdot \mathbf{F}(\mathbf{R}_m, \omega). \end{aligned} \quad (5.12)$$

This is the central result of this section. It is an exact Lippmann-Schwinger equation and it describes the resonant scattering off and emission by guest atoms inside an inhomogeneous

dielectric. It has an undisturbed term, a scattering and a source term. The elements of equation (5.12) must still be explained: the operator  $\mathbf{E}^{(0)}(\mathbf{r}, \omega)$  is the electric field in the absence of the atoms, with both the (+) and the (−) parts; the atomic source operators  $\mathbf{S}_m(\omega)$  are vectors and have the form

$$\mathbf{S}_m(\omega) \equiv \hat{\boldsymbol{\mu}}_m S_m(\omega) \equiv \hat{\boldsymbol{\mu}}_m \left( \frac{-i\mu_m \omega^2}{\varepsilon_0 c^2} \right) \left[ \frac{b_m(0)}{\omega - \Omega_m} + \frac{b_m^\dagger(0)}{\omega + \Omega_m} \right], \quad (5.13)$$

where  $\hat{\boldsymbol{\mu}}_m$  denotes the unit vector in the direction of the atomic dipole moment  $\boldsymbol{\mu}_m$ . Notice that  $\mathbf{S}_m$  features the atomic creation and annihilation operators at the initial time zero: in quantum optics, the atomic variables can not be completely integrated out in an “all-light” picture. Spontaneous emission will be described in terms of these source operators in section 5.3.

The optical potentials produced by the atoms are dyadics and are given by

$$\mathbf{V}_m(\omega) \equiv \hat{\boldsymbol{\mu}}_m V_m(\omega) \hat{\boldsymbol{\mu}}_m \equiv \hat{\boldsymbol{\mu}}_m \left( \frac{\mu_m^2 \omega^2}{\hbar \varepsilon_0 c^2} \right) \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \hat{\boldsymbol{\mu}}_m. \quad (5.14)$$

The potentials  $\mathbf{V}_m(\omega)$  are tensorial instead of isotropic because the atoms can not be polarized in directions perpendicular to their dipole moments. Both the sources and the potentials have resonances at frequencies  $\pm\Omega_m$ . The potentials  $V_m(\omega)$  can be rewritten as  $-(\omega/c)^2 \alpha_{Bm}(\omega)$ , with the frequency-dependent “bare polarizability” [69] defined as

$$\alpha_{Bm}(\omega) = \frac{2\mu_m^2 \Omega_m}{\hbar \varepsilon_0 (\Omega_m^2 - \omega^2)}. \quad (5.15)$$

The bare polarizabilities are real (except on resonance) and change sign when going through their resonances at  $\Omega_m$ ; the resonances are infinitely sharp because all possible nonradiative decay processes are neglected; a polarizability is called “bare” precisely because its resonance is not radiatively broadened.

The last undefined factor in the Lippmann-Schwinger equation (5.12) is the dyadic quantity  $\mathbf{K}$  which is given by

$$\mathbf{K}(\mathbf{r}, \mathbf{r}', \omega) \equiv c^2 \sum_{\lambda} \frac{\mathbf{f}_{\lambda}(\mathbf{r}) \mathbf{f}_{\lambda}^*(\mathbf{r}')}{(\omega^2 - \omega_{\lambda}^2)} \frac{\omega_{\lambda}^2}{\omega^2} \quad (5.16)$$

In the denominator, the frequency  $\omega$  is assumed to contain an infinitesimally small positive imaginary part  $i\eta$ , so that  $\mathbf{K}$  has an imaginary part whenever  $\omega$  coincides with a mode frequency  $\omega_{\lambda}$ . The dyadic  $\mathbf{K}$  can be interpreted once a few Green functions of the inhomogeneous dielectric are introduced. This is done in some detail in appendix C. The relevant results of this appendix are repeated here: the (full) Green tensor  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$  of the inhomogeneous dielectric is the solution of the wave equation

$$-\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) + \varepsilon(\mathbf{r})(\omega/c)^2 \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') \mathbf{I}, \quad (5.17)$$

where the right-hand side is the ordinary Dirac delta function times the unit tensor. The generalized transverse Green function  $\mathbf{G}^T$  is the solution of

$$-\nabla \times \nabla \times \mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) + \varepsilon(\mathbf{r})(\omega/c)^2 \mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) = \bar{\delta}_{\varepsilon}^T(\mathbf{r}', \mathbf{r}). \quad (5.18)$$

The source term  $\bar{\delta}_\varepsilon^T$  at the right-hand side is the transpose (denoted by a bar) of the generalized transverse delta function (4.12). The generalized transverse Green function  $\mathbf{G}^T$  has an expansion

$$\mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) = c^2 \sum_{\lambda} \frac{\mathbf{f}_{\lambda}(\mathbf{r}) \mathbf{f}_{\lambda}^*(\mathbf{r}')}{\omega^2 - \omega_{\lambda}^2}, \quad (5.19)$$

in terms of the (complex) normal modes of the homogeneous wave function of the dielectric; the frequency  $\omega$  has an infinitesimally small positive imaginary part. With these definitions (5.17)–(5.19), the dyadic  $\mathbf{K}$  (5.16) can be rewritten as

$$\mathbf{K}(\mathbf{r}, \mathbf{r}', \omega) = c^2 \sum_{\lambda} \frac{\mathbf{f}_{\lambda}(\mathbf{r}) \mathbf{f}_{\lambda}^*(\mathbf{r}')}{\omega^2 - \omega_{\lambda}^2} - (c/\omega)^2 \sum_{\lambda} \mathbf{f}_{\lambda}(\mathbf{r}) \mathbf{f}_{\lambda}^*(\mathbf{r}') \quad (5.20)$$

$$= \mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) - \frac{1}{(\omega/c)^2 \varepsilon(\mathbf{r}')} \bar{\delta}_\varepsilon^T(\mathbf{r}, \mathbf{r}'). \quad (5.21)$$

It consists of the generalized transverse Green function (5.19) and a term proportional to the generalized transverse delta function  $\bar{\delta}_\varepsilon^T$  (4.24). Both terms are medium-dependent. Note that  $\mathbf{K}$  is generalized transverse in its variable  $\mathbf{r}$ . In particular, the second term in (5.21) is not a longitudinal term. Because of its transversality property,  $\mathbf{K}$  is not equal to the total Green function (5.17). Nevertheless, both for free space and also for the dielectric, the definition of the longitudinal Green function [see appendix C, Eq. (C.3)] can be used to rewrite the dyadic  $\mathbf{K}$  as

$$\mathbf{K}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \frac{1}{\varepsilon(\mathbf{r})(\omega/c)^2} \delta(\mathbf{r} - \mathbf{r}') \mathbf{I}. \quad (5.22)$$

According to this identity, the dyadic  $\mathbf{K}$  that shows up in the Lippmann-Schwinger equation (5.12) for the Field only differs from the full Green function of the medium when its two position arguments  $\mathbf{r}$  and  $\mathbf{r}'$  coincide. The delta function term in (5.22) is a self-interaction term that arises in the multipolar formalism: the dipole couples not only to the electric field but also to its own polarization field through the  $-\boldsymbol{\mu} \cdot \mathbf{D}/[\varepsilon_0 \varepsilon(\mathbf{R})]$  interaction that was derived in the previous chapter. Although it is different from  $\mathbf{G}$ , the quantity  $\mathbf{K}$  will also be called a Green function in the following.

All the elements of the Lippmann-Schwinger equation (5.12) for the Field have now been explained. Solutions for the Field  $\mathbf{F}(\mathbf{r}, \omega)$  will be given in later sections. Once such a solution is known, also the vector potential (4.25a) and the magnetic field can be determined right away in terms of the Field, as it turns out by taking the appropriate linear combinations of creation and annihilation operators (5.11a) and (5.11b):

$$\begin{aligned} \mathbf{A}(\mathbf{r}, \omega) &\equiv \sum_{\lambda} \sqrt{\frac{\hbar}{2\varepsilon_0 \omega_{\lambda}}} \left[ a_{\lambda}(\omega) \mathbf{f}_{\lambda}(\mathbf{r}) + a_{\lambda}^{\dagger}(\omega) \mathbf{f}_{\lambda}^*(\mathbf{r}) \right] \\ &= \mathbf{A}^{(0)}(\mathbf{r}, \omega) + \frac{1}{i\omega} \sum_m \mathbf{G}^T(\mathbf{r}, \mathbf{R}_m, \omega) \cdot [\mathbf{S}_m(\omega) + \mathbf{V}_m(\omega) \cdot \mathbf{F}(\mathbf{R}_m, \omega)]. \end{aligned} \quad (5.23)$$

Here the vector potential is propagated by the generalized transverse Green function  $\mathbf{G}^T$ , rather than by  $\mathbf{K}$ , as in the equation (5.12) for the Field. The magnetic field is the curl of the vector potential.

In previous chapters, Lippmann-Schwinger equations were derived in “all-light” formalisms that started with a given optical potential as a perturbation. Here instead, the approach starts one level deeper with a dynamical system of light and matter. The dynamics of the atoms were integrated out in order to mimic an all-light formalism as much as possible. The optical potential  $\mathbf{V}_m$  is output rather than input.

A new feature (as compared to Lippmann-Schwinger equations in previous chapters) is that in equation (5.12) the atoms not only are point scatterers (potentials), but also point sources. Both appear as two sides of the same coin in one equation. The Field is a sum of two terms that will be called  $\mathbf{F}_1$  and  $\mathbf{F}_2$ . The Field  $\mathbf{F}_1$  contains the undisturbed Field  $\mathbf{E}^{(0)}$  plus the Field that is scattered by the guest atoms;  $\mathbf{F}_2$  is the source Field associated with the radiation of guest dipoles that are excited at the initial time zero. Expectation values of  $\mathbf{F}_1$  only depend on the initial state of the electromagnetic field, whereas expectation values involving  $\mathbf{F}_2$  depend only on the initial atomic state.

### 5.2.3 Volume-integrated dipole field

As a first application and test of the formalism, the (operator) relation between the Field and the polarization field of the atoms is now determined. The polarization field  $\mathbf{P}_{gm}$  (4.61) associated with guest atom  $m$  in the dipole approximation is

$$\mathbf{P}_{gm}(\mathbf{r}, t) = \delta(\mathbf{r} - \mathbf{R}_m) \mathbf{P}_{gm}(t) = \delta(\mathbf{r} - \mathbf{R}_m) \boldsymbol{\mu}_m [b_m(t) + b_m^\dagger(t)]. \quad (5.24)$$

With the help of the equations (5.10a), (5.10b), and the definitions of the source fields (5.13) and potentials (5.14), the polarization field in frequency space can be related to other operators as

$$\mathbf{P}_{gm}(\omega) = - \left( \frac{\varepsilon_0 c^2}{\omega^2} \right) [ \mathbf{S}_m(\omega) + \mathbf{V}_m(\omega) \cdot \mathbf{F}(\mathbf{R}_m, \omega) ]. \quad (5.25)$$

There exists therefore a simple relationship between the Field and the polarization fields [use Eq. (5.12)]

$$\mathbf{F}(\mathbf{r}, \omega) = \mathbf{E}^{(0)}(\mathbf{r}, \omega) - \frac{\omega^2}{\varepsilon_0 c^2} \sum_{m=1}^N \mathbf{K}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{P}_{gm}(\omega). \quad (5.26)$$

Now assume that the sources are in free space. The volume-integral will now be determined of the Field over a small sphere (denoted by  $\odot$ ) containing only the source at  $\mathbf{R}_m$ , at its center. This integral is important in many situations, for example when studying local-field effects. The only contribution to the integral that survives when making the radius of the sphere smaller, stems from the source  $m$  itself. The integral is determined by the properties of the free-space Green function  $\mathbf{K}_0(\mathbf{r}, \mathbf{R}_m, \omega)$  at positions  $\mathbf{r}$  close to  $\mathbf{R}_m$ . For convenience, the form (5.21) of the function  $\mathbf{K}$  will be used in the following discussion rather than (5.22). The transverse Green function  $\mathbf{G}_0^T(\mathbf{r} - \mathbf{R}_m, \omega)$  in  $\mathbf{K}_0$  has a vanishing contribution to the integral, since its pole goes as  $|\mathbf{r} - \mathbf{R}_m|$  at short distances [69]. The dipole part of the transverse delta function (C.5) has a vanishing solid-angle-integral and

does not contribute either. What remains is the delta-function part of the transverse delta function, which gives the radius-independent result

$$\int_{\odot} d\mathbf{r} \mathbf{F}(\mathbf{r}, \omega) = \frac{2}{3\varepsilon_0} \mathbf{P}_{gm}(\omega). \quad (5.27)$$

Now the subtlety becomes important that the Field  $\mathbf{F}$  is equal to the electric field  $\mathbf{E}$  everywhere except at the positions of the guest atoms. The right-hand side of equation (5.27) is therefore not equal to the volume-integrated electric field. With the definitions of the displacement field (4.46) and the Field (5.2), one obtains the following relation between the volume integrals of the electric-field operator and the polarization-field operator around an atom

$$\int_{\odot} d\mathbf{r} \mathbf{E}(\mathbf{r}, \omega) = -\frac{1}{3\varepsilon_0} \mathbf{P}_{gm}(\omega). \quad (5.28)$$

The static and classical version of this result is presented for example in Jackson's book [4], where a delta-function contribution to the static dipole field is added by hand for consistency; likewise, in the theory for classical point scatterers by De Vries *et al.* [69], it is stated that a delta-function contribution must be added by hand to the longitudinal Green function  $\mathbf{G}_0^L$  to ensure that the classical analogy of the "sum rule" (5.28) holds. In contrast, Eq. (5.28) was found here as an operator relation without adding any terms by hand. The sum rule (5.28) could only be found by using the fact that a dipole couples to the field  $-\mathbf{D}(\mathbf{R})/[\varepsilon_0\varepsilon(\mathbf{R})]$  rather than to minus the electric field: in the present calculation there is only one correct answer to the dipole coupling controversy (see section 4.6).

There is nothing truly quantum mechanical about the sum rule (5.28). In a classical canonical theory, one would find the same conjugate fields (4.50), dipole coupling (4.62) and Green function  $\mathbf{K}$ . However, a canonical formalism is usually by-passed in classical optics. It is then assumed that a classical dipole couples to the classical electric field and furthermore that light propagates from a source according to the Green function  $\mathbf{G}$  rather than  $\mathbf{K}$ . In appendix C it is shown that  $\mathbf{G}_0$  naturally has the correct delta-function term in its longitudinal part. There is no need to add terms by hand in order to derive Eq. (5.28) classically.

Incidentally, in obtaining the result (5.28), the definition of the longitudinal Green function was not used: the dipole couples to a transverse field so that  $\mathbf{G}_0^L$  does not come into play. Still, the same volume-integrated electric field could be found when carrying out the calculations using the form (5.22) for  $\mathbf{K}$ . The latter form does depend on the definition of the longitudinal Green function.

## 5.3 Single-atom properties altered by the medium

An atom in a group of atoms in an inhomogeneous dielectric will have different properties as compared to free space, because of the dielectric and because of the other atoms. In this section the effect of the medium on the individual atoms will be considered. The next step, in section 5.4, will be to study some effects that the medium-modified atoms can have on each other. Thus, the results of the present section are necessary ingredients for the many-atom problem.

### 5.3.1 Light emitted by a point source

Assume that in the dielectric there is only one guest atom present with dipole moment  $\boldsymbol{\mu}$  and transition frequency  $\Omega$ . The effect of the medium on the scattering and emission properties of the atom can be found by solving the equation (5.12). The Green function  $\mathbf{K}$  in that equation can not be replaced by the Green function  $\mathbf{G}$ , since the delta function in  $\mathbf{K}$  plays a role especially when considering multiple scattering off a single atom. The Field can be solved exactly from equation (5.12) by successive iterations

$$\mathbf{F} = [\mathbf{E}^{(0)} + \mathbf{K} \cdot \mathbf{S}] + \mathbf{K} \cdot \mathbf{V} \cdot [\mathbf{E}^{(0)} + \mathbf{K} \cdot \mathbf{S}] + \mathbf{K} \cdot \mathbf{V} \cdot \mathbf{K} \cdot \mathbf{V} \cdot [\mathbf{E}^{(0)} + \mathbf{K} \cdot \mathbf{S}] + \dots \quad (5.29)$$

In this equation,  $\mathbf{K}$  and  $\mathbf{V}$  are classical quantities, whereas  $\mathbf{F}$ ,  $\mathbf{E}^{(0)}$ , and  $\mathbf{S}$  are operators. The infinite series can be summed, much the same as was done in chapters 2 and 3 for light scattering by plane scatterers:

$$\mathbf{F}(\mathbf{r}, \omega) = \mathbf{F}_1(\mathbf{r}, \omega) + \mathbf{F}_2(\mathbf{r}, \omega), \quad \text{where} \quad (5.30)$$

$$\mathbf{F}_1(\mathbf{r}, \omega) = \mathbf{E}^{(0)}(\mathbf{r}, \omega) + \mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{E}^{(0)}(\mathbf{R}, \omega) \quad (5.31)$$

$$\mathbf{F}_2(\mathbf{r}, \omega) = \mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{S}(\omega) + \mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{K}(\mathbf{R}, \mathbf{R}, \omega) \cdot \mathbf{S}(\omega). \quad (5.32)$$

In this exact solution for the Field,  $\mathbf{F}_1$  describes light that is either undisturbed or scattered by the guest atoms; the source Field  $\mathbf{F}_2$  can be related to spontaneous emission by the atoms. The solutions of  $\mathbf{F}_2$  and  $\mathbf{F}_1$  will be discussed here and in section 5.3.2, respectively.

The single-atom T-matrix is

$$\mathbf{T}(\omega) = \hat{\boldsymbol{\mu}} T(\omega) \hat{\boldsymbol{\mu}} = \hat{\boldsymbol{\mu}} \left[ \frac{V(\omega)}{1 - \hat{\boldsymbol{\mu}} \cdot \mathbf{K}(\mathbf{R}, \mathbf{R}, \omega) \cdot \hat{\boldsymbol{\mu}} V(\omega)} \right] \hat{\boldsymbol{\mu}} \quad (5.33)$$

and clearly, it depends on the atomic position inside the inhomogeneous dielectric. The source Field (5.32) can be rewritten as

$$\mathbf{F}_2(\mathbf{r}, \omega) = \frac{\mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{S}(\omega)}{1 - \hat{\boldsymbol{\mu}} \cdot \mathbf{K}(\mathbf{R}, \mathbf{R}, \omega) \cdot \hat{\boldsymbol{\mu}} V(\omega)}, \quad (5.34)$$

with  $\mathbf{S}(\omega)$  as defined in Eq. (5.13). The time dependence can be calculated of the Field at the position  $\mathbf{r}$  due to the presence of the source at  $\mathbf{R}$ , by doing the inverse Laplace transformation:

$$\mathbf{F}_2(\mathbf{r}, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \frac{(\omega^2 - \Omega^2) \mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{S}(\omega)}{\omega^2 - \Omega^2 - \hat{\boldsymbol{\mu}} \cdot \mathbf{K}(\mathbf{R}, \mathbf{R}, \omega) \cdot \hat{\boldsymbol{\mu}} [2\mu^2 \omega^2 \Omega / (\hbar \varepsilon_0 c^2)]}, \quad (5.35)$$

This integral in general can not be evaluated further without the explicit knowledge of the Green function. Light can go from the source at  $\mathbf{R}$  to a detector at position  $\mathbf{r}$  following several paths, dependent on the geometry of the dielectric. The Green function contains information about all possible paths. For example, if an atom near a mirror emits light, the light can either directly go to the detector or first reflect off the mirror. Each path has its own retardation time due to the finite speed of light. Light from all these paths interferes at position  $\mathbf{r}$ .

One can only say something about the the amplitude at position  $\mathbf{r}$  of the field emitted by the source when the Green function is known, but the decay rate of the source can be found already. By making a pole approximation in the frequency poles of Eq. (5.35), one will find that the source-Field operator will decay exponentially in time. Exponential decay is only found because all orders in the atom-radiation coupling were taken into account. The (amplitude) spontaneous-decay rate is

$$\Gamma/2 \equiv -\hat{\boldsymbol{\mu}} \cdot \text{Im } \mathbf{K}(\mathbf{R}, \mathbf{R}, \Omega) \cdot \hat{\boldsymbol{\mu}} \left( \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \right). \quad (5.36)$$

The decay rate of the intensity of the field is  $\Gamma$ . From the definition of  $\mathbf{K}$  in Eq. (5.16) it is clear that this rate is a nonnegative quantity. The delta function term in  $\mathbf{K}$  (5.22) is real, so that the imaginary part of  $\mathbf{K}$  is equal to the imaginary part of the Green function  $\mathbf{G}$ . More specifically, since the longitudinal Green function  $\mathbf{G}^L$  (C.3) is real, the decay rate is proportional to the imaginary part of only  $\mathbf{G}^T$ . The decay rate depends both on the atom's position and on its orientation inside the inhomogeneous dielectric. For free space, the imaginary part of  $\mathbf{G}_0^T(\mathbf{R}, \mathbf{R}, \Omega)$  is equal to  $-\Omega/(6\pi c)\mathbf{I}$  [see Eq. (C.7)]. This immediately gives the familiar free-space spontaneous-decay rate  $\Gamma_0 = \mu^2 \Omega^3 / (3\pi \hbar \varepsilon_0 c^3)$ .

The decay rate is accompanied by a shift of the oscillation frequency  $\Omega$  by an amount

$$\Delta' \equiv \hat{\boldsymbol{\mu}} \cdot \text{Re } \mathbf{K}(\mathbf{R}, \mathbf{R}, \Omega) \cdot \hat{\boldsymbol{\mu}} \left( \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \right). \quad (5.37)$$

For two reasons, this frequency shift is infinitely large even in free space. The first reason is that the delta function term  $\delta(\mathbf{r} - \mathbf{R}_m)\mathbf{I}/[\varepsilon(\mathbf{r})(\omega/c)^2]$  in the expression (5.22) for  $\mathbf{K}$  diverges when  $\mathbf{r}$  is equal to  $\mathbf{R}$ . The term was already interpreted as a self-interaction and it seems to be medium-dependent through the factor  $\varepsilon(\mathbf{r})$ . This is not the case, however, if an empty-cavity model is assumed so that the dielectric function is equal to 1 at the position of guest atom (see section 4.4.3). In that model, the self-interaction causes a medium-independent infinite shift. The other reason why the frequency shift (5.37) diverges is well known for free space: the Green function  $\mathbf{G}(\mathbf{r}, \mathbf{R}, \omega)$  diverges when  $\mathbf{r}$  approaches  $\mathbf{R}$  (see appendix C for the explicit form of  $\mathbf{G}_0$ ). By a procedure called mass renormalisation, the combined radiative shift in free space becomes finite [16]. There is yet another reason why infinite line shifts could appear: in the present formalism the dipole approximation was made. For consistency and in order to prevent possible infinite line shifts due to the dipole approximation, the mode expansion of the Green function should be cut off at high frequencies  $\omega_\lambda = 2\pi c/a$ , where  $a$  is the size of the atom. This cutoff will not be shown explicitly in the following. From now on it is assumed that  $\Omega$  is the observable atomic frequency in vacuum, being the sum of the “bare frequency” and the radiative shift in vacuum. Inside a dielectric, the atomic frequency can get a finite shift with respect to the vacuum value  $\Omega$ , a shift that is given by

$$\Delta = \hat{\boldsymbol{\mu}} \cdot \text{Re} [ \mathbf{G}(\mathbf{R}, \mathbf{R}, \Omega) - \mathbf{G}_0(\mathbf{R}, \mathbf{R}, \Omega) ] \cdot \hat{\boldsymbol{\mu}} \left( \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \right), \quad (5.38)$$

and it clearly depends on the atomic position and dipole orientation. The expression (5.38) has already been used in section 3.5 for the calculation of line shifts near a plane scatterer.

Effectively, after redefining the transition frequency  $\Omega$ , the factor  $\mathbf{K}$  in the denominator of the expression (5.33) for the single-atom T-matrix must be replaced by  $\mathbf{G}(\mathbf{R}, \mathbf{R}, \omega) - \text{Re } \mathbf{G}_0(\mathbf{R}, \mathbf{R}, \omega)$ .

The position-dependent radiative shifts are a mechanism of inhomogeneous broadening of the detected light. Experimentally, it is hard to single out the radiative shift (a photonic effect) from electronic line shifts (due to changes in the atomic wave functions inside the medium). Electronic shifts usually dominate inhomogeneous broadening. Electronic line shifts are absent in the empty-cavity local-field model, but in practice the model is not expected to work so well that radiative shifts can be detected. Electronic effects are minimized if the guest atoms are in the gas phase (inside an air-sphere crystal for example). Their positions are not fixed then and collision and Doppler broadening mechanisms come into play, unless the atoms are strongly cooled. For these reasons it will be hard to observe a position-dependent radiative shift in a dielectric.

### 5.3.2 Light scattered by a point scatterer

Spontaneous emission of point sources in the dielectric has now been discussed by studying the source field  $\mathbf{F}_2$ . There is also the other contribution  $\mathbf{F}_1$  to the Field, consisting of the undisturbed electric field and a scattered field:

$$\mathbf{F}_1(\mathbf{r}, \omega) = \mathbf{E}^{(0)}(\mathbf{r}, \omega) + \mathbf{K}(\mathbf{r}, \mathbf{R}, \omega) \cdot \mathbf{T}(\omega) \cdot \mathbf{E}^{(0)}(\mathbf{R}, \omega). \quad (5.39)$$

In this equation, the atom appears as a point scatterer with an internal resonance [69] in the optical potential  $V(\omega)$  that appears in the definition (5.33) of the T-matrix. The Field  $\mathbf{F}_1$  feels both the inhomogeneous dielectric and the optical potential due to the atom. The scattered Field term has poles at  $\pm\omega_\lambda$  and also at the poles of the T-matrix. The first poles describe direct light scattering off the guest atoms, whereas the poles of the T-matrix describe resonance fluorescence of light that is first captured by the atom and subsequently is re-emitted. These two processes can be disentangled by separating the frequency poles (straightforward, but not spelled out here), followed by an inverse Laplace transformation. As for the source Field in Eq. (5.35), the frequency poles are simple but the full time-dependence can only be calculated when an explicit form of the Green function is known. For the part of  $\mathbf{F}_1$  featuring the annihilation operators, one finds

$$\begin{aligned} \mathbf{F}_1^{(+)}(\mathbf{r}, t) = \mathbf{E}^{(0)(+)}(\mathbf{r}, t) &+ \sum_{\lambda} \frac{\mu^2}{2\pi\hbar\epsilon_0 c^2} \sqrt{\frac{\hbar\omega_\lambda}{2\epsilon_0}} a_{\lambda}^{(0)}(0) \mathbf{f}_{\lambda}(\mathbf{r}) \cdot \hat{\boldsymbol{\mu}} \times \\ &\int_{-\infty}^{\infty} d\omega e^{-i\omega t} \left\{ \begin{aligned} &- \frac{2\Omega_s \omega_\lambda^2 \mathbf{K}(\mathbf{r}, \mathbf{R}, \Omega_s) \cdot \hat{\boldsymbol{\mu}}}{\Omega_s^2 - \omega_\lambda^2 + \Gamma^2/4 - i\omega_\lambda \Gamma} \cdot \frac{1}{\omega - \omega_\lambda} \\ &+ \frac{\Omega_s^2 \mathbf{K}(\mathbf{r}, \mathbf{R}, \Omega_s) \cdot \hat{\boldsymbol{\mu}}}{\Omega_s - \omega_\lambda - i\Gamma/2} \cdot \frac{1}{\omega - \Omega_s + i\Gamma/2} \\ &+ \frac{\Omega_s^2 \mathbf{K}^*(\mathbf{r}, \mathbf{R}, \Omega_s) \cdot \hat{\boldsymbol{\mu}}}{\Omega_s + \omega_\lambda + i\Gamma/2} \cdot \frac{1}{\omega + \Omega_s + i\Gamma/2} \end{aligned} \right\} \quad (5.40) \end{aligned}$$

Here,  $\Omega_s$  is the shifted frequency defined as  $\Omega + \Delta$ ;  $\Delta$  and  $\Gamma$  were defined in Eqs. (5.38) and (5.36), respectively. The negative-frequency part  $\mathbf{F}^{(-)}$  of the Field equals  $[\mathbf{F}^{(+)}]^\dagger$ .

The three terms between curly brackets in (5.40) correspond to three optical processes. The first term describes elastic light scattering by the guest atom inside the inhomogeneous dielectric; the second term has an exponentially decaying time dependence and corresponds to resonance fluorescence; finally, the third term is also exponentially decaying and moreover it is nonresonant because of the  $(\Omega_s + \omega_\lambda)$  in the denominator. This third term corresponds to an utterly improbable process that one could call anti-resonance fluorescence; in a rotating-wave approximation this process would disappear. After neglecting this third term, all the  $\omega$ -poles in the integral (5.40) have positive real parts, so that (5.40) can be called the positive-frequency part of the Field  $\mathbf{F}_1$ .

As a further identification of the elastic-scattering term, the free-space far-field differential cross section  $d\sigma/d\Omega$  will now be determined. (Confusion between the solid angle  $\Omega$  and the frequency  $\Omega$  should not arise.) The Green function is known and the frequency integrals in (5.40) can be performed. For elastic scattering,  $d\sigma/d\Omega$  is given by  $|r|^2 \langle I_s \rangle / \langle I_p \rangle$ , where  $\mathbf{R} = 0$  is assumed;  $\langle I_s \rangle$  and  $\langle I_p \rangle$  are the expectation values of the scattered intensity at position  $\mathbf{r}$  and of the pump intensity, respectively [15]. Assume monochromatic polarized pump light with wave vector  $\mathbf{k}_p$ , frequency  $\omega_p = k_p c$  and polarisation direction  $\boldsymbol{\varepsilon}_p$  in a coherent state  $|\alpha_p\rangle$ . The pump light intensity is  $\hbar\omega_p c |\alpha_p|^2$ , where Eq. (5.1) was used for the intensity operator. The average intensity of the scattered light (with polarisation direction  $\boldsymbol{\varepsilon}_s$ ) can be found by inserting the first term between curly brackets of Eq. (5.40) also into Eq. (5.1) and taking the expectation value. This leads to the differential cross section

$$d\sigma/d\Omega = \frac{(\boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}_p)^2 (\boldsymbol{\mu} \cdot \boldsymbol{\varepsilon}_s)^2}{4\pi^2 \hbar^2 \varepsilon_0^2 c^4} \cdot \frac{\Omega_s^2 \omega_p^4}{(\Omega_s^2 - \omega_p^2 + \Gamma^2/4)^2 + \omega_p^2 \Gamma^2}. \quad (5.41)$$

This cross section is proportional to  $\omega_p^4$  in the Rayleigh limit  $\omega_p/\Omega_s \ll 1$  and it is independent of the pump frequency in the Thomson limit  $\omega_p/\Omega_s \gg 1$ . The present formalism gives the correct behavior in both these limiting cases, because the rotating-wave approximation was not made. At resonance, the present T-matrix formalism is better than the Kramers-Heisenberg dispersion formula [15, 85] when applied to elastic scattering, because radiative broadening is included here.

The fluorescent light [the second term between curly brackets in Eq. (5.40)] has the same position-dependent emission rates  $\Gamma(\mathbf{R}, \Omega)$  (5.36) and line shifts  $\Delta(\mathbf{R}, \Omega)$  (5.38) as found for spontaneous emission in the source Field. In the fluorescence process the guest atom is excited by light of frequency  $\omega_p$  in a pulse that passes the atom, after which the atom decays exponentially, following the atomic dynamics. The shapes of the incoming pulse and the elastically scattered pulse depend on the interference and time-dependence of many optical modes. Obviously, a more detailed description of such a pulsed experiment is required, but it will not be given here.

### 5.3.3 Single atom as a point source in “homogeneous” dielectric

The general results for single atoms of section 5.3.1 will now be applied in a special case: the spontaneous-emission rate and radiative shift of a guest atom inside a homogeneous dielectric will be calculated, both with and without local-field corrections. As in chapter 4, the guest atoms are described microscopically and the dielectric is characterized

macroscopically by a homogeneous dielectric constant  $\varepsilon(\mathbf{r}) = \varepsilon = n^2$ . The dielectric can not be really homogeneous on the microscopic level. This point was already discussed qualitatively in section 4.4.3 by considering the Hamiltonian for the guest atoms.

First assume that the dielectric is homogeneous everywhere. Let  $\mathbf{G}_{\text{macro}}$  be the Green function for the homogeneous medium with  $\varepsilon$  a constant everywhere. Compare the wave equation (C.1) for this Green function when  $\varepsilon(\mathbf{r}) = \varepsilon$  with the free-space wave equation for  $\mathbf{G}_0$  with  $\varepsilon(\mathbf{r}) = 1$ . It follows that  $\mathbf{G}_{\text{macro}}(\mathbf{r}, \omega)$  is equal to  $\mathbf{G}_0(\mathbf{r}, n\omega)$ . This identity also holds for the transverse (C.7) and longitudinal (C.8) parts separately:

$$\begin{aligned}\mathbf{G}_{\text{macro}}^{\text{T}}(\mathbf{r}, \omega) &= -\frac{\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi(n\omega/c)^2 r^3} - \frac{e^{in\omega r/c}}{4\pi r} [P(in\omega r/c)\mathbf{I} + Q(in\omega r/c)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}] \\ \mathbf{G}_{\text{macro}}^{\text{L}}(\mathbf{r}, \omega) &= \frac{\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi(n\omega/c)^2 r^3} + \frac{\delta(\mathbf{r})}{3(n\omega/c)^2} \mathbf{I},\end{aligned}\quad (5.42)$$

with the function  $P(z)$  defined as  $\equiv (1 - z^{-1} + z^{-2})$  and  $Q(z)$  as  $(-1 + 3z^{-1} - 3z^{-2})$  (see appendix C). The spontaneous-emission rate  $\Gamma_{\text{macro}}$  that one then finds with Eq. (5.36) is  $n\Gamma_0$ , the refractive index times the free-space rate  $\Gamma_0$ . The same rate can be found using Fermi's golden rule by assuming that the dipole couples to a field consisting of plane waves with frequencies  $\omega_{\mathbf{k}} = kc/n$ . The rate  $n\Gamma_0$  follows from the assumption that the dielectric is homogeneous everywhere. The assumption entails that the local field felt by the guest atom is equal to the macroscopic Maxwell Field.

What field does a guest atom feel? It was found in section 4.4.5 that a dipole couples to the Field  $\mathbf{F}$  rather than to the electric field. This formal result is not under debate here. The question is rather whether  $\mathbf{F}$  can be written in terms of an expansion of plane-wave modes with frequencies  $\omega_{\mathbf{k}} = kc/n$  or that modifications are required in the vicinity of the guest atom. There are several competing models that give different predictions for the local field (see [94, 95] for an overview). Here the empty-cavity model will be considered. Some of the other models require a (partly) microscopic description of the dielectric beyond the present macroscopic theory. The best known other model is the full-cavity (or virtual-cavity) model [4]. Recent experiments [91] and microscopic calculations [94] suggest that the empty-cavity model gives correct predictions when the guest atom cuts out a sphere that is larger than the size of the constituents of the dielectric.

In the empty-cavity model [11], the dielectric is considered homogeneous with a dielectric constant  $\varepsilon$ , except in a sphere with radius  $R_{\odot}$  much smaller than the optical wavelength. The guest atom sits in the center of this tiny sphere with dielectric function equal to 1. It was found in section 4.4.3 that the absence of huge chemical shifts of the energy levels of impurity atoms requires a negligible overlap of  $[\varepsilon(\mathbf{r}) - 1]$  and the relevant wave functions of the guest atom. The empty-cavity model satisfies this requirement if  $R_{\odot}$  is larger than the size of the atom. The optical modes in the empty-cavity model are modified plane waves and they were already calculated in [11]. The reason to present the calculation here in a slightly different way is to exploit the real-space representation of the free-space Green function, and to be able to use intermediate results later in section 5.5 about superradiance.

The empty sphere in the homogeneous dielectric can be described by the optical po-

tential

$$\mathbf{V}_\odot \equiv \theta(R_\odot - |\mathbf{r}|)(\varepsilon - 1)(\omega/c)^2 \mathbf{I}, \quad (5.43)$$

which acts as a local perturbation in the wave equation for the modes. Here,  $\theta(x)$  is the step-function. A macroscopic plane-wave mode  $\mathbf{f}_{\mathbf{k},\sigma}^{\text{macro}}(\mathbf{r})$  is equal to  $\sigma_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{r})/\sqrt{\varepsilon}$ . The guest atom couples to the Field at  $\mathbf{r} = 0$ . In the origin, the mode function  $\mathbf{f}_{\mathbf{k}\sigma}^\odot$  of the dielectric-with-hole satisfies the Lippmann-Schwinger equation

$$\mathbf{f}_{\mathbf{k}\sigma}^\odot(0) = \sigma_{\mathbf{k}} + (\varepsilon - 1)(\omega/c)^2 \int_\odot d\mathbf{r}' \mathbf{G}_{\text{macro}}(-\mathbf{r}', \omega) \cdot \mathbf{f}_{\mathbf{k}\sigma}^\odot(\mathbf{r}'). \quad (5.44)$$

The Green function in this equation is the sum of the transverse and longitudinal Green functions (5.42). Now since  $kR_\odot \ll 1$  is assumed, the short-distance behavior of the Green function  $\mathbf{G}_{\text{macro}}$  is important. In that limit,  $\mathbf{G}^T$  has an integrable divergence, as follows from Eq. (5.42). The integral over the cavity volume of  $\mathbf{G}^T$  can therefore be neglected. (For the same reason, local modifications of scalar waves due to a tiny empty sphere would be negligible because the divergence of  $g_0(\mathbf{r}) = -\exp(i\omega r)/(4\pi r)$  is integrable as well.) On the other hand, Eq. (5.42) also shows that  $\mathbf{G}^L$  has a non-integrable  $r^{-3}$  divergence and a delta function term as well. In the Born approximation and in all higher-order approximations one finds that only the delta-function term of  $\mathbf{G}^L$  gives a finite contribution; the other term gives zero after the angular integration, because the cavity is assumed spherical. Therefore the solution of the mode function at the position of the guest atom is

$$\mathbf{f}_{\mathbf{k}\sigma}^\odot(0) = \left( \frac{3\varepsilon}{2\varepsilon + 1} \right) \mathbf{f}_{\mathbf{k},\sigma}^{\text{macro}}(0). \quad (5.45)$$

The factor  $\mathcal{L}_e \equiv 3\varepsilon/(2\varepsilon + 1)$  is called the empty-cavity local-field factor. From the mode expansion (5.19) one sees that the imaginary part of the generalized-transverse part of the Green function  $\mathbf{G}^\odot(0, 0)$  is enhanced by a factor  $\mathcal{L}_e^2$  as compared to  $\mathbf{G}_{\text{macro}}(\mathbf{R}, \mathbf{R})$ ; the spontaneous-emission rates should be enhanced by  $\mathcal{L}_e^2$  as well [11].

Line shifts in a medium are also caused by interaction of the atom with the local field rather than with the macroscopic field. Both the free-space Green function (C.7) and the Green function (5.46) of the dielectric have diverging real parts when both  $\mathbf{r}$  and  $\mathbf{r}'$  approach the atomic position  $\mathbf{R}$ . However, the difference between the two diverging terms is zero in that limit. With Eq. (5.38) one finds that there is no additional radiative shift for light emitted in a spherical empty cavity in a homogeneous dielectric.

Not only spontaneous-emission rates are changed when modes  $\mathbf{f}_\odot$  must be used rather than the plane-wave modes  $\mathbf{f}_{\text{macro}}$ . In principle, the generalized transverse Green function (5.19), the generalized transverse delta function (4.12) and the longitudinal Green function (C.3) are all changed. Since the polarizability of the tiny empty sphere (guest atom excluded) inside the dielectric is negligible, the differences between  $\mathbf{f}_\odot(\mathbf{r})$  and  $\mathbf{f}_{\text{macro}}(\mathbf{r})$  as described in Eq. (5.44) can be neglected for positions  $\mathbf{r} \neq \mathbf{R}$ . Consequently, one can say that

$$\mathbf{G}_\odot(\mathbf{r}, \mathbf{r}', \omega) = [\mathcal{L}_e]^n \mathbf{G}_{\text{macro}}(\mathbf{r}, \mathbf{r}', \omega), \quad (5.46)$$

where  $n$  is the total number of position arguments of the Green function that coincides with an atomic position  $\mathbf{R}$ .

## 5.4 Several atoms as point sources and scatterers

In section 5.3 it was studied how scattering and emission rates of single atoms can be influenced by their dielectric surroundings. Only photonic effects were taken into account and electronic effects were neglected. In the present section it is studied how the medium-modified atoms can influence each other. Analogous to section 5.3, here the atomic wave functions are assumed not to overlap each other and to be unaffected by the dielectric. The general method to solve the Lippmann-Schwinger equation in this more complicated situation is outlined here. In section 5.5, the formalism developed in this section will be applied to the problem of superradiance of two guest atoms inside an inhomogeneous dielectric medium.

The Lippmann-Schwinger equation (5.12) for the Field was already solved for only one guest atom. With more atoms present, solutions of the Field can be found iteratively from the implicit equation (5.12). The Field is known exactly and explicitly when the  $N$ -atom T-matrix in the inhomogeneous dielectric can be found. The total Field is the sum of  $\mathbf{F}_1$  and  $\mathbf{F}_2$ . The scattered Field  $\mathbf{F}_1$  solves the homogeneous and the source field  $\mathbf{F}_2$  the inhomogeneous wave equation:

$$\mathbf{F}_1(\mathbf{r}, \omega) = \mathbf{E}^{(0)}(\mathbf{r}, \omega) + \sum_{m,n=1}^N \mathbf{K}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{T}_{mn}^{(N)}(\omega) \cdot \mathbf{E}^{(0)}(\mathbf{R}_n, \omega). \quad (5.47)$$

The  $N$ -atom T-matrix can indeed be calculated explicitly and has the form

$$\mathbf{T}_{mn}^{(N)}(\omega) = \hat{\boldsymbol{\mu}}_m T_{mn}^{(N)}(\omega) \hat{\boldsymbol{\mu}}_n = \hat{\boldsymbol{\mu}}_m T_m(\omega) M_{mn}^{-1}(\omega) \hat{\boldsymbol{\mu}}_n, \quad (5.48)$$

where the  $N \times N$  matrix  $\mathbf{M}(\omega)$  is defined as

$$M_{ij}(\omega) = [\delta_{ij} - (1 - \delta_{ij}) \hat{\boldsymbol{\mu}}_i \cdot \mathbf{G}(\mathbf{R}_i, \mathbf{R}_j, \omega) \cdot \hat{\boldsymbol{\mu}}_j T_j(\omega)]. \quad (5.49)$$

The basic ingredients of this T-matrix are the Green function  $\mathbf{G}$  of the inhomogeneous dielectric and the atomic frequencies and dipole moments. With these, the position-dependent single-atom T-matrices  $T_m(\omega)$  and the dipole-dipole interaction strengths  $\hat{\boldsymbol{\mu}}_m \cdot \mathbf{G}(\mathbf{R}_m, \mathbf{R}_n, \omega) \cdot \hat{\boldsymbol{\mu}}_n$  can be determined. In the matrix  $M$  the Green function  $\mathbf{K}$  (5.22) should have shown up instead of the  $\mathbf{G}$ , if not the factor  $(1 - \delta_{ij})$  made them interchangeable.

The scattering of light by the atoms makes that light does not propagate as described by  $\mathbf{G}$  anymore. Equations (5.48) and (5.49) neatly sum up infinitely many scattering events which are not described by  $\mathbf{G}$ . The occurrence of  $\mathbf{G}$  in Eq. (5.49) signifies that light does propagate as described by  $\mathbf{G}$  in between the scattering off one atom and the next one. This is a more general scattering situation than in chapters 2 and 3, where the scatterers were assumed to be embedded inside free space, and scattering events were connected by the scalar and dyadic free-space Green functions  $g_0$  and  $\mathbf{G}_0$ , respectively.

By using the same iteration that led to the expression (5.47) for  $\mathbf{F}_1$ , the source-field  $\mathbf{F}_2$  can also be expressed in terms of the T-matrix:

$$\mathbf{F}_2(\mathbf{r}, \omega) = \sum_{m=1}^N \mathbf{K}^{(N)}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{S}_m(\omega). \quad (5.50)$$

Here,  $\mathbf{K}^{(N)}$  is the Green function of the inhomogeneous dielectric including the  $N$  atoms, also including self-interactions of the atoms:

$$\mathbf{K}^{(N)}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{K}(\mathbf{r}, \mathbf{r}', \omega) + \sum_{m,n=1}^N \mathbf{K}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{T}_{mn}^{(N)}(\omega) \cdot \mathbf{K}(\mathbf{R}_n, \mathbf{r}', \omega). \quad (5.51)$$

At positions  $\mathbf{r}$  and  $\mathbf{r}'$  different from one of the guest atoms,  $\mathbf{K}^{(N)}(\mathbf{r}, \mathbf{r}', \omega)$  is equal to what one would call the total Green function  $\mathbf{G}^{(N)}$  of both the dielectric and its guests. The solution (5.50) shows that the source-field  $\mathbf{K}^{(N)}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{S}_m(\omega)$  that emanates from atom  $m$  is influenced by the positions, orientations, dipole moments and resonance frequencies of the  $(N-1)$  other atoms from which the light can scatter.

Equation (5.47) is an explicit solution for the operator of the scattered electric field. The present formalism can therefore be used to study nonclassical light scattered off a collection of point scatterers in all directions. (Quantum optics of random dielectrics with a discrete number of modes was studied in [46].) The atomic positions can be considered as random variables that sometimes have to be averaged over [68].

The scattered field  $\mathbf{F}_1$  and the source Field  $\mathbf{F}_2$  have been treated separately, but it is important to notice that each scatterer is also source: many elastic scatterers in free space together form an (on average) homogeneous dielectric with complex dielectric function [68]; the (averaged) source Field operator will serve as the quantum noise source that keeps the commutation relations of the (average) field operators time-independent. (More about quantum noise in dielectrics in chapter 6.)

The source field  $\mathbf{F}_2$  formed by the guest atoms is decaying in time. This section ends with discussing the different situation of a continuous field produced by a pumped point source. Unlike the dynamics of the guest atoms, the dynamics of the pumped source is not included in the Hamiltonian (5.4a). Suppose that we have the same inhomogeneous dielectric as before with the same guest atoms as before. The novelty is that at position  $\mathbf{r}_p$  light is emitted from a continuous source  $\mathbf{S}_p$ :

$$\mathbf{S}_p(\mathbf{r}, t) = \delta(\mathbf{r} - \mathbf{r}_p) [\mathbf{s}_p e^{-i\omega_p t} + \mathbf{s}_p^* e^{+i\omega_p t}]. \quad (5.52)$$

This source is the analogue of the sources (5.13) of the guest atoms. The pumped source emits light at a frequency  $\omega_p$ . Assume that light, once emitted, does not return to the pumped source. In that case, the influence of the source on the total Green function of the dielectric plus guest atoms can be neglected. If the source starts radiating at time zero, then the emitted field is

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= \int d\mathbf{r}' \int_0^t dt' \mathbf{K}^{(N)}(\mathbf{r}, \mathbf{r}', t - t') \cdot \mathbf{S}_p(\mathbf{r}', t') \\ &= \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} \mathbf{K}^{(N)}(\mathbf{r}, \mathbf{r}_p, \omega) \cdot \left[ \frac{\mathbf{s}_p}{\omega - \omega_p} + \frac{\mathbf{s}_p^*}{\omega + \omega_p} \right]. \end{aligned} \quad (5.53)$$

In the last equality, it was assumed that the time  $t$  was long enough that  $\sin[(\omega - \omega_p)t]/(\omega - \omega_p)$  could be taken to be a delta function [15]. As before, the frequency  $\omega$  in the denominators of Eq. (5.53) is understood to have an infinitesimally small positive imaginary part  $+i\eta$ .

The source (5.52) and field (5.53) can be either classical quantities or operators. So (5.52) could be a classical source with an amplitude  $s_p$ . In principle, this amplitude could be analyzed into more elementary properties, but here it is treated as a given quantity. Or the source (5.52) could be an operator, just like Eq. (5.13). Then the quantum mechanical state of the source (coherent, squeezed, etcetera) should be a given quantity here, so that expectation values involving (5.53) can be determined.

## 5.5 Two-atom superradiance in inhomogeneous medium

### 5.5.1 Motivation

The general results of section 5.4 for point scatterers and point sources will now be applied to two identical atoms which are positioned in an inhomogeneous dielectric. Two identical atoms in free space much less than a wavelength apart either cooperatively decay with twice the single-atom decay rate, or do not decay at all [45, 125, 126]. These superradiance effects can be viewed as caused by retarded multiple-scattering interactions, see [127–129] and references therein. Lifetime changes of individual atom pairs as a function of their distance were measured only recently [119]; for two  $\text{Ba}^+$  ions that emit at a wavelength of 493 nm and for well-defined separations  $|\mathbf{R}|$  around 1.5  $\mu\text{m}$ , subradiant and superradiant lifetime effects of less than  $\pm 2\%$  were observed. High-resolution interference experiments of one and two atoms in the presence of a (distant) mirror were reported recently [130].

The spatial variations in single-atom spontaneous-emission rates can be much larger than the lifetime changes due to superradiance cited above: inside photonic crystals lifetimes can be dramatically different for positions less than half an optical wavelength apart (see chapters 2 and 3). Moreover, interatomic interactions will also change inside a dielectric. In studies of superradiance of  $N$  atoms in photonic crystals, the atoms are assumed to be confined in a volume  $\lambda^3$ , but the inevitable inhomogeneity felt by the atoms is neglected, or treated as a random variable [23, 24]. However, a volume of optical dimensions is too large to assume identical local densities of states everywhere inside, yet it is too small to consider the LDOS a random variable. On the wavelength scale of light, the LDOS in a photonic crystal is an engineered rather than a random variable. It is therefore an interesting question how two-atom superradiance is modified in an inhomogeneous dielectric, in the close vicinity of a mirror for example, inside an optical cavity, or inside a photonic crystal. In the following calculations, the dielectric constant  $\varepsilon(\mathbf{r})$  will be left arbitrary (but real), which encompasses the interesting special cases just mentioned.

In the literature on superradiance, the atoms are usually modelled as two-level atoms. Here they will be modelled as quantum harmonic oscillators in their ground states or first excited states, for which the point source formalism of section 5.4 is applicable. It is known that not only two- or more-level atoms exhibit superradiance, but so do quantum harmonic oscillators. Superradiance has even been observed for classical oscillators such as tuning forks in acoustics [45]. These models have in common that the atoms (or oscillators) interact with a common field that is influenced by the radiation reactions of all nearby atoms together.

### 5.5.2 Calculation of the two-atom source Field

Assume that the two atoms have identical transition frequencies  $\Omega$  and dipole moments  $\mu = |\mu|$ ; their dipole orientations  $\hat{\mu}_1$  and  $\hat{\mu}_2$  need not be identical. The source field of this two-atom system can be calculated from equation (5.50) of section 5.4:

$$\mathbf{F}_2(\mathbf{r}, \omega) = \mathbf{K}^{(2)}(\mathbf{r}, \mathbf{R}_1, \omega) \cdot \mathbf{S}_1(\omega) + \mathbf{K}^{(2)}(\mathbf{r}, \mathbf{R}_2, \omega) \cdot \mathbf{S}_2(\omega). \quad (5.54)$$

The goal is now to calculate the Green function  $\mathbf{K}^{(2)}$  of the dielectric including the guest atoms, in terms of the properties of the medium and of the individual atoms. Once that has been accomplished, the time-dependence of the source Field will be determined as a function of the initial excited state of the atoms. In this section 5.5.2 these results will be derived in rather condensed form; their interpretation is postponed until section 5.5.3.

According to Eq. (5.51), the Green function  $\mathbf{K}^{(2)}$  is known once the T-matrix  $T^{(2)}$  (5.48) is determined;  $T^{(2)}$  can be found by inverting the  $2 \times 2$  matrix  $M^{(2)}$  (5.49), in which the single-atom  $T$  matrices occur that are given in equation (5.33) and the Green function  $\mathbf{K}$  of the dielectric in (5.22). The calculation therefore starts with the matrix

$$M^{(2)} = \begin{pmatrix} 1 & -\hat{\mu}_1 \cdot \mathbf{G}(\mathbf{R}_1, \mathbf{R}_2, \omega) \cdot \hat{\mu}_2 T_2 \\ -\hat{\mu}_2 \cdot \mathbf{G}(\mathbf{R}_2, \mathbf{R}_1, \omega) \cdot \hat{\mu}_1 T_1 & 1 \end{pmatrix}. \quad (5.55)$$

Let  $J_{12}$  be short-hand notation for  $\beta \hat{\mu}_1 \cdot \mathbf{G}(\mathbf{R}_1, \mathbf{R}_2, \omega) \cdot \hat{\mu}_2$ , with  $\beta$  defined as  $\mu^2 \omega^2 / (\hbar \varepsilon_0 c^2)$ . Then  $J_{21} = J_{12}$  and the two-atom T-matrix is

$$\mathbf{T}^{(2)} = \frac{1}{1 - T_1 J_{12}^2 T_2 / \beta^2} \begin{pmatrix} \hat{\mu}_1 \hat{\mu}_1 T_1 & \hat{\mu}_1 \hat{\mu}_2 T_1 J_{12} T_2 / \beta \\ \hat{\mu}_2 \hat{\mu}_1 T_2 J_{12} T_1 / \beta & \hat{\mu}_2 \hat{\mu}_2 T_2 \end{pmatrix}. \quad (5.56)$$

Each of the four matrix elements of  $\mathbf{T}^{(2)}$  is a dyadic of the same type as the single-atom T-matrix (5.33). The frequency dependence in the Green function  $\mathbf{K}(\mathbf{r}, \mathbf{R}_1, \omega)$  will be dropped and it will be further abbreviated as  $\mathbf{K}(\mathbf{r}1)$ ; other terms are written in similar short-hand notation. The Green function  $\mathbf{K}^{(2)}(\mathbf{r}1)$  can be written with Eq. (5.51) as

$$\begin{aligned} \mathbf{K}^{(2)}(\mathbf{r}1) &= \mathbf{K}(\mathbf{r}1) \cdot \left[ \mathbf{I} + \mathbf{T}_{11}^{(2)} \cdot \mathbf{K}(11) + \mathbf{T}_{12}^{(2)} \cdot \mathbf{K}(21) \right] \\ &+ \mathbf{K}(\mathbf{r}2) \cdot \left[ \mathbf{T}_{21}^{(2)} \cdot \mathbf{K}(11) + \mathbf{T}_{22}^{(2)} \cdot \mathbf{K}(21) \right]. \end{aligned} \quad (5.57)$$

It is convenient to split off the vector character from the source terms  $\mathbf{S}_j$  (5.13) by rewriting them as  $\hat{\mu}_j S_j$ . Use Eq. (5.56) to rewrite the T-matrix elements of  $\mathbf{T}^{(2)}$  in terms of the single-atom T-matrices. The first one of the two parts of the source Field (5.54) is associated with light initially residing in atom 1. This part can be written in terms of single-atom properties as

$$\mathbf{K}^{(2)}(\mathbf{r}1) \cdot \mathbf{S}_1 = \left( \frac{1 + T_1 X_1 / \beta}{1 - T_1 J_{12}^2 T_2 / \beta^2} \right) [\mathbf{K}(\mathbf{r}1) \cdot \hat{\mu}_1 + \mathbf{K}(\mathbf{r}2) \cdot \hat{\mu}_2 T_2 J_{21} / \beta] S_1. \quad (5.58)$$

Here,  $X_1$  is defined as

$$X_1 = \Omega_1(\omega) - \Omega = \Delta_1(\omega) - i\Gamma_1(\omega)/2, \quad (5.59)$$

and  $\Omega_1 = \Omega_1(\omega)$  is the complex single-atom resonance frequency in the medium.  $X_2$  and  $\Omega_2$  are defined analogously for the second atom. When  $X_1$  equals  $X_2$ , the atoms are said to be placed at equivalent positions in the dielectric. The source field has now been expressed in terms of the T-matrices of the individual atoms, but is is rewarding to break up the T-matrices in parts that depend on the medium alone and parts that depend on the atoms. The equation (5.58) is written in more elementary quantities as

$$\mathbf{K}^{(2)}(\mathbf{r}_1) \cdot \mathbf{S}_1 = \frac{(\omega^2 - \Omega^2)S_1 \left[ \mathbf{K}(\mathbf{r}_1) \cdot \hat{\boldsymbol{\mu}}_1 ((\omega^2 - \Omega^2) - 2\Omega X_2) + \mathbf{K}(\mathbf{r}_2) \cdot \hat{\boldsymbol{\mu}}_2 2\Omega J_{12} \right]}{(\omega^2 - \Omega^2)^2 - 2\Omega(X_1 + X_2)(\omega^2 - \Omega^2) + 4\Omega^2(X_1 X_2 - J_{12}^2)}. \quad (5.60)$$

The denominator carries the important information about the resonance frequencies  $\Omega_{\pm}(\omega)$  of the two-atom system:

$$\begin{aligned} \Omega_{\pm}^2 &= \Omega^2 + 2\Omega \left[ \frac{1}{2}(X_1 + X_2) \pm \sqrt{\left(\frac{X_1 - X_2}{2}\right)^2 + J_{12}^2} \right] \\ &\simeq \left[ \Omega_1 + \Omega_2 \pm \sqrt{\left(\frac{\Omega_1 - \Omega_2}{2}\right)^2 + J_{12}^2} \right]^2. \end{aligned} \quad (5.61)$$

Actually, there are four resonance frequencies, both at  $\Omega_{\pm}(\omega)$  and at  $-\Omega_{\pm}(\omega)$ . The approximation that was made in the second line of equation (5.61) is good when the resonance frequencies change little due to the electromagnetic coupling with the dielectric, that is when  $|\Omega_{\pm}(\Omega) - \Omega|/\Omega$  is much smaller than one. In high-quality optical cavities the matter-light coupling is so strong that this approximation breaks down and more resonance frequencies appear [131]. (Another example will be given in section 5.6.) Now rewrite Eq. (5.60) as a sum over individual first-order frequency poles. After using the definition (5.13) of the source operator  $\mathbf{S}_1$ , one has

$$\begin{aligned} \mathbf{K}^{(2)}(\mathbf{r}_1) \cdot \mathbf{S}_1 &= \sum_{\pm} \left( \frac{-i\mu\omega^2}{4\varepsilon_0 c^2 \Omega_{\pm}} \right) \left[ \mathbf{K}(\mathbf{r}_1) \cdot \hat{\boldsymbol{\mu}}_1 (1 \pm \sin \alpha) \pm \mathbf{K}(\mathbf{r}_2) \cdot \hat{\boldsymbol{\mu}}_2 \cos \alpha \right] \\ &\times \left[ (\omega + \Omega)b_1(0) + (\omega - \Omega)b_1^{\dagger}(0) \right] \left[ \frac{1}{\omega - \Omega_{\pm}} - \frac{1}{\omega + \Omega_{\pm}} \right]. \end{aligned} \quad (5.62)$$

A (complex) angle  $\alpha$  has been introduced which measures the inhomogeneity of the medium as felt by the two-atom system, in comparison with the atom-atom interaction:

$$\sin \alpha \equiv \frac{(\Omega_1 - \Omega_2)}{\sqrt{(\Omega_1 - \Omega_2)^2 + 4J_{12}^2}}; \quad \cos \alpha \equiv \frac{2J_{12}}{\sqrt{(\Omega_1 - \Omega_2)^2 + 4J_{12}^2}}. \quad (5.63)$$

The frequency dependence of  $\alpha$  is not shown explicitly. At equivalent positions such as in free space, the angle  $\alpha$  is zero.

The time-dependence of the source field can now be calculated with an inverse Laplace transformation. The relative change of  $\Omega_{\pm}(\omega)$  as compared to  $\Omega$  was assumed small already. It is therefore natural to make a pole approximation at this point and to replace in

the denominators  $\Omega_{\pm}(\omega)$  by  $\Omega_{\pm} \equiv \Omega_{\pm}(\Omega)$ . Notice that the positive-frequency poles in Eq. (5.62) have negligible contributions to terms proportional to  $b_1^\dagger$ ; similarly, negative-frequency poles hardly contribute to terms involving the annihilation operator  $b_1(0)$ . The total source Field  $\mathbf{F}_2(\mathbf{r}, t)$  is the Field (5.62) that originates from the initial excitation of the atom labelled 1, accompanied by the source Field that originally came from the second atom:

$$\mathbf{F}_2(\mathbf{r}, t) = \sum_{\pm} \{ \mathbf{L}_{1\pm}(\mathbf{r}, t) b_1(0) + \mathbf{L}_{2\pm}(\mathbf{r}, t) b_2(0) + \text{H.c.} \}. \quad (5.64)$$

The vectors  $\mathbf{L}_{1\pm}$  are defined as

$$\begin{aligned} \mathbf{L}_{1\pm}(\mathbf{r}, t) \equiv & \frac{-i\mu\Omega_{\pm}^2}{2\varepsilon_0 c^2} \int_{-\infty}^{\infty} d\omega \frac{e^{i\omega t}}{\omega - \Omega_{\pm}} \times \\ & [ \mathbf{K}(\mathbf{r}, \mathbf{R}_1, \Omega_{\pm}) \cdot \hat{\boldsymbol{\mu}}_1 (1 \pm \sin \alpha_{\pm}) \pm \mathbf{K}(\mathbf{r}, \mathbf{R}_2, \Omega_{\pm}) \cdot \hat{\boldsymbol{\mu}}_2 \cos \alpha_{\pm} ], \end{aligned} \quad (5.65)$$

where  $\alpha_{\pm}$  is written as short-hand notation for  $\alpha(\Omega_{\pm})$ . The vectors  $\mathbf{L}_{2\pm}$  can be found by interchanging the indices 1 and 2 in the right-hand side of Eq. (5.65), which also causes a sign change in  $\sin \alpha$  (5.63). The explicit form (5.64) of the time-dependent source field of the two-atom system is the central result of this rather dense section. It is this result that will be interpreted and used in the next section.

### 5.5.3 Effects of two-atom superradiance

In the previous section 5.5.2, the source field (5.64) was calculated of two near-by resonant atoms inside an inhomogeneous dielectric. Equation (5.1) shows that this is enough information to calculate the time-dependent intensity of the emitted light at an arbitrary detector position. Suppose that atom 1 is brought in its (first) excited state at time  $t = 0$  and atom 2 is not excited, so that  $|\Psi(t = 0)\rangle = b_1^\dagger(0)|0\rangle$ . Then from Eqs. (5.1) and (5.64) it follows that at time  $t$ , the expectation value of the intensity operator is given by

$$\langle I(\mathbf{r}, t) \rangle = 2\varepsilon_0 c | \mathbf{L}_{1+}(\mathbf{r}, t) + \mathbf{L}_{1-}(\mathbf{r}, t) |^2. \quad (5.66)$$

The time-dependent intensity that passes at  $\mathbf{r}$  is a complicated interference pattern of light coming from four sources, a fast(er) and a (more) slowly decaying source at  $\mathbf{R}_1$ , and also a fast and a slow source at the atomic position  $\mathbf{R}_2$ . There is interference of light, even though initially all the light stems from the excited first atom. The situation is different in Young's double-slit experiment where there is only interference when both slits are light sources [15]. In the present case, non-interacting slits are replaced by interacting atoms; a photon is exchanged between the atoms until it is finally emitted.

When the atoms are far apart, then  $J_{12}$  tends to zero and the two resonance frequencies  $\Omega_{\pm}$  (5.61) are simply the two single-atom frequencies  $\Omega_1$  and  $\Omega_2$  with their medium-dependent radiative shifts  $\Delta_{1,2}$  and decay rates  $\Gamma_{1,2}$ . When initially only the first atom is excited, the probability that the photon once emitted is captured by the far-away second atom is negligible. Interference will not be observed. Only when both well-separated sources are excited initially, will interference exist.

In the other extreme situation, for atoms (almost) on top of each other, one has in free space that  $X_1 = X_2 \equiv X^{(0)}$  and the decay rates are proportional to  $-\text{Im}[X^{(0)} \pm J_{12}^{(0)}]$ . The decay rates are  $2\Gamma_0$  and 0 when the interatomic distances go to zero, since then  $J_{12}$  approaches  $X^{(0)}$ . Half of the excited-state population remains trapped in the two atoms. For the two atoms in the dielectric one finds from (5.61) similar results in the limit  $|\mathbf{R}_1 - \mathbf{R}_2| \rightarrow 0$ : the local densities of states felt by the two atoms become almost equal;  $-\text{Im}\Omega_+$  approaches twice the single-atom amplitude decay rate, whereas  $-\text{Im}\Omega_-$  has the limiting value zero.

The intermediate situations of inequivalent atoms at moderate distances is more interesting where neither  $(\Omega_1 - \Omega_2)$  nor  $J_{12}$  is negligible. In the expression (5.61) for the resonance frequencies and in the angle  $\alpha$  (5.63), a driving term and a detuning can be discerned. The driving term is  $J_{12}$  and it signifies how important the one atom is as a light source for the other. The term  $(\Omega_1 - \Omega_2)/2$  acts as a detuning: larger local differences felt by the identical atoms make the resonant transfer of a photon between them less probable. The driving term and the detuning have the same physical origin and can not be changed independently. By bringing the atoms very close, they will be tuned better and interact stronger at the same time.

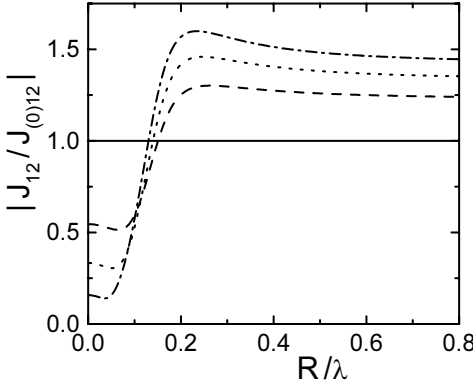
The effect of detuning can be clarified further by studying the part  $\mathbf{F}_2^{\text{2nd}}(\mathbf{r}, t)$  of the total source field  $\mathbf{F}_2(\mathbf{r}, t)$  that is finally emitted from the initially unexcited second atom. This part consists of a faster and a slower source term:

$$\mathbf{F}_2^{\text{2nd}}(\mathbf{r}, t) = \frac{-i\mu b_1(0)}{2\epsilon_0 c^2} \sum_{\pm} \int d\omega \frac{\pm\omega^2 \cos \alpha_{\pm} \mathbf{G}(\mathbf{r}, \mathbf{R}_2, \Omega_{\pm}) \cdot \boldsymbol{\mu}_2 e^{-i\omega t}}{\omega - \Omega_{\pm}} + \text{H.c.} \quad (5.67)$$

For far-apart atoms without detuning, the cosines are equal to 1 but the two source terms cancel because both resonance frequencies  $\Omega_{\pm}$  are equal; the sum of the two terms in (5.67) can also be neglected when the atoms are so far apart or so far detuned that  $|J_{12}|$  is much smaller than  $|\Omega_1 - \Omega_2|/2$ : the terms do not cancel, but are negligible separately since  $\cos \alpha_{\pm} \ll 1$ . In both situations, a negligible fraction of the light is finally emitted from atom 2 when initially only atom 1 is excited. The single-atom emission (5.35) is found back in both situations. This leads to an important observation: the cooperative effects of the two identical atoms will decrease when their local optical environments differ more. The detuning of the atoms consists not only of differences in line shifts; differences in local densities of states of the medium add to the (complex) detuning. Since near mirrors or in photonic crystals the LDOS varies considerably on the sub-wavelength scale, local differences in LDOS felt by the two atoms will be the cause of considerable detuning and consequently a diminishing of the cooperative effects.

## 5.6 Two-atom superradiance in “homogeneous” dielectrics

It has been known since a long time that the interaction between two resonant atoms is influenced by their dielectric environment [132]. The formalism of this chapter will now be used to study how two-atom superradiance is modified inside a nondispersive homogeneous dielectric medium with  $\epsilon(\mathbf{r}) = \epsilon = n^2$ . The results of two-atom superradiance of

**Figure 5.1:**

Absolute values of the dipole-dipole interaction  $J_{12}(R)$  of parallel dipoles, as a function of their scaled distance  $R/\lambda$  in homogeneous nondispersive dielectrics with refractive index  $n$ .  $|J_{12}(R)|$  is given relative to the free-space value  $|J_{(0)12}(R)|$ . The curves are based on Eq. (5.68). Solid line:  $n = 1.0$ ; dashed line:  $n = 1.5$ ; dotted line:  $n = 2.0$ ; dot-dashed line:  $n = 3.0$ .

section 5.5 were obtained for inhomogeneous dielectrics, and the homogeneous dielectric is a limiting case covered in that theory. It is worthwhile to calculate the effects of the homogeneous medium before going back to the inhomogeneous case in section 5.7.

The atoms are separated by a distance  $R \equiv |\mathbf{R}_1 - \mathbf{R}_2|$ . Since the dielectric is homogeneous, the positions of the two atoms are equivalent and detuning due to differences in the local environment is absent. When local-field effects would be neglected, one finds that not only emission rates (as in section 5.3.3) but also atomic dipole-dipole interactions can be described by the free-space Green function  $\mathbf{G}_0$  with its argument  $\omega$  replaced by  $n\omega$  (5.42). In a more precise model, local-field effects are taken into account. The same local modifications (5.45) that modify the local density of states felt by a single atom in an empty cavity, also show up in the dipole-dipole interaction  $J_{12}$  between two atoms. Assume that the dipole moments  $\boldsymbol{\mu}_{1,2}$  are equal in magnitudes  $\mu$  and orientations and assume further that  $\hat{\boldsymbol{\mu}}_{1,2} \cdot \mathbf{R} = 0$  (“ $\Sigma$ -configuration”). With Eq. (5.46) one finds the dipole-dipole interaction

$$J_{12}(n, \mathbf{R}, \Omega) = - \left( \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \right) \frac{e^{in\Omega R/c}}{4\pi R} \left\{ 1 + \frac{ic}{n\Omega R} - \frac{c^2}{n^2 \Omega^2 R^2} \right\} \left( \frac{3n^2}{2n^2 + 1} \right)^2. \quad (5.68)$$

The absolute value of this quantity, relative to the free-space value, is given as a function of interatomic distance in figure 5.1, for several refractive indices of the dielectric.

The figure shows that  $J_{12}$  depends sensitively on both distance and refractive index. In the figure, the interatomic distance is scaled relative to the free-space wavelength  $\lambda \equiv 2\pi c/\Omega$  of the emitted light. The overall local-field correction  $\mathcal{L}_e^2$  is larger than unity for  $n > 1.0$ . Still, in the near field ( $R/\lambda \lesssim 0.1$ ),  $|J_{12}|$  is smaller than in free space and more suppressed for higher refractive indices; there is a cross-over regime ( $0.1 \lesssim R/\lambda \lesssim 0.3$ ) where the dipole-dipole interaction is climbing up to more than the free-space value and the faster so for larger  $n$ , until a maximum is attained in the relative interaction; in the far field, the only relative enhancement of the interaction originates from the local-field term  $\mathcal{L}_e^2$ . This far-field limit is to be compared with the recent measurements on two-atom superradiance in free space, where the atoms were separated by about three optical wavelengths [119].

Figure 5.1 shows that superradiance is strongly influenced by the medium, at least

for the dipole orientations chosen. For two guest atoms in a gas, relative orientations  $\hat{\mathbf{R}}$  are not fixed and the average interaction is described by the angle-averaged Green function  $\mathcal{L}_e^2(\mathbf{G}_0(\mathbf{R}, n\Omega))_{av.}$ , see Eq. (C.9). The angle-averaged dipole interaction then has the same absolute value as in free space, except for the local-field factor. The averaging procedure does not apply to individual atom pairs in a solid, which have fixed positions and dipole orientations.

In figure 5.2(a), line shifts  $\text{Re}(\Omega_+ - \Omega)$  are compared for free space with  $n = 1.0$  and for a dielectric with  $n = 2.0$ . The shifts are divided by the free-space amplitude decay-rate  $\Gamma_0/2$ . The plots are based on Eqs. (5.61) and (5.68). From the figure, it is clear that the line shifts oscillate as a function of distance, with a larger amplitude in the dielectric and with a period that is a factor  $n$  faster than in free space. Shifts corresponding to  $\Omega_-$  are not shown, but they can be found easily since the extra shift in the dielectric is zero (section 5.3.3) so that  $\text{Re}(\Omega_- - \Omega)$  equals minus  $\text{Re}(\Omega_+ - \Omega)$ . For distances  $R/\lambda \lesssim 0.1$ , line shifts become much larger than  $\Gamma_0/2$ . At those short distances,  $J_{12}$  can be approximated by

$$J_{12}(n, \mathbf{R}) = \left( \frac{\mu^2}{4\pi\hbar\epsilon_0 R^3} \right) \frac{1}{n^2} \left( \frac{3n^2}{2n^2 + 1} \right)^2. \quad (5.69)$$

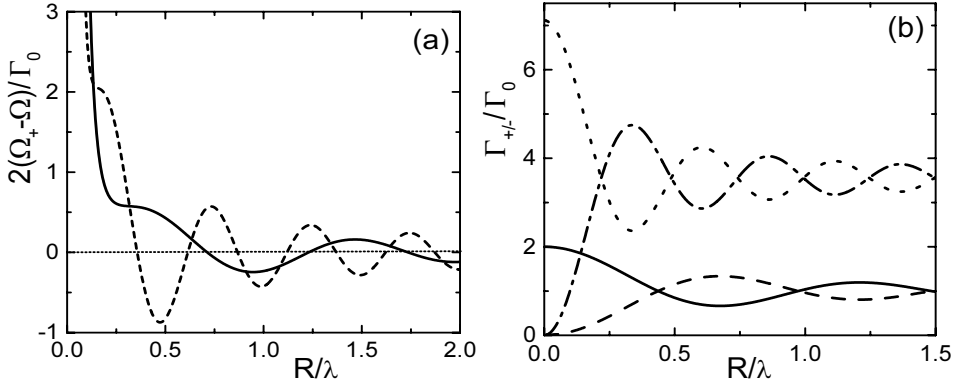
The phase factor due to the finite speed of light could be neglected and the frequency dependence dropped out completely. The medium reduces the interaction by the factor  $n^{-2}$  but enhances it by the square of the empty-cavity local-field factor.

Since the dipole-dipole interaction  $\hbar J_{12}$  has no frequency-dependence for very close atoms, in the realm of chemical physics it is often modelled as a static interaction term in an effective Hamiltonian for the atoms only [133]. It is a well-known effect in molecular assemblies like  $J$ -aggregates [134] and in biological light-harvesting systems [135] that emitted and absorbed light have frequencies that are shifted significantly (the so-called “ $J$ -shift”) with respect to the single-molecule frequency  $\Omega$ . Equation (5.69) is in qualitative agreement with those observations. However, the observed shifts occur in systems where typical separations are of the order of one nanometer. For these very small distances, (5.69) can not be expected to be valid quantitatively, because the assumption made in Eq. (5.61) breaks down that the shifts are small compared to  $\Omega$ . Furthermore, the assumption that each atom is embedded independently in a spherical empty cavity inside the host dielectric will also break down in this limit.

In figure 5.2(b), decay rates  $\Gamma_{\pm} \equiv -2\text{Im}\Omega_{\pm}$  are compared for  $n = 1.0$  and  $n = 2.0$ . The decay rates oscillate as a function of interatomic distance, and faster for larger  $n$ . The limiting value for large distance is the single-particle decay-rate  $n\mathcal{L}_e^2\Gamma_0$ . Twice this value is the short-distance limit of  $\Gamma_+$ , while  $\Gamma_-$  tends to zero.

## 5.7 Discussion: superradiance in photonic crystals

The homogeneous dielectric of section 5.6 was a special case of the results obtained in section 5.5. Now reconsider the general case of superradiance in inhomogeneous dielectrics. As seen in section 5.5.2, two-atom superradiance sensitively depends on the competition between their medium-induced detuning and their dipole-dipole interaction. The (complex) detuning  $(\Omega_1 - \Omega_2)/2$  of two atoms with identical transition frequencies  $\Omega$  depends

**Figure 5.2:**

Line shifts (a) and emission rates (b) of a two-atom system as a function of interatomic distance. In (a): the solid line is the shift  $\text{Re}(\Omega_+ - \Omega)$  for  $n = 1.0$ , scaled to  $\Gamma_0/2$ ; the dashed line is the same plot for  $n = 2.0$ . In (b): the solid line represents  $\Gamma_+/\Gamma_0$  for  $n = 1.0$ ; the dashed line depicts  $\Gamma_-/\Gamma_0$  for  $n = 1.0$ ; the dotted line is  $\Gamma_+/\Gamma_0$  for  $n = 2.0$ ; finally, the dot-dashed line shows  $\Gamma_-/\Gamma_0$  for  $n = 2.0$ .

on the radiative shifts  $\Delta(\mathbf{R}_{1,2})$  (5.38) and on the decay rates  $\Gamma(\mathbf{R}_{1,2})$  (5.36):

$$\Omega_1 - \Omega_2 = \Delta(\mathbf{R}_1) - \Delta(\mathbf{R}_2) - i [\Gamma(\mathbf{R}_1) - \Gamma(\mathbf{R}_2)]/2. \quad (5.70)$$

Superradiance will only occur when  $|\Omega_1 - \Omega_2|/2$  is smaller than the absolute value of the dipole-dipole interaction

$$J_{12}(\mathbf{R}_1, \mathbf{R}_2, \Omega) = \frac{\mu^2 \Omega^2}{\hbar \epsilon_0 c^2} \hat{\mu}_1 \cdot \left\{ c^2 \sum_{\lambda} \frac{\mathbf{f}_{\lambda}(\mathbf{R}_1) \mathbf{f}_{\lambda}^*(\mathbf{R}_2)}{\Omega^2 - \omega_{\lambda}^2} - c^2 \sum_{\lambda} \frac{\mathbf{f}_{\lambda}(\mathbf{R}_1) \mathbf{f}_{\lambda}^*(\mathbf{R}_2)}{\Omega^2} \right\} \cdot \hat{\mu}_2, \quad (5.71)$$

where Eqs. (5.20) and (5.22) were used. This interaction will be discussed briefly. The first term in (5.71) is proportional to the transverse Green function  $\mathbf{G}^T(\mathbf{R}_1, \mathbf{R}_2, \Omega)$  (5.19). It is frequency-dependent and is called the “resonant dipole-dipole interaction” (RDDI). The second term of (5.71) is proportional to the longitudinal Green function  $\mathbf{G}^L(\mathbf{R}_1, \mathbf{R}_2, \Omega)$  (C.3). This second term is the “static dipole-dipole interaction”, or SDDI. Both the resonant and the static dipole-dipole interactions are described in terms of the generalized-transverse modes of the dielectric.

The resonant dipole-dipole interaction in Eq. (5.71) can be enhanced when inserting the atoms into a dielectric (a cavity, for example) with mode frequencies  $\omega_{\lambda}$  equal to  $\Omega$ . Detailed calculations of the RDDI in optical cavities were performed in [120–122]. Experimental proof of the enhancement of the RDDI inside an optical cavity was given in [123]. In photonic crystals, mode functions with frequencies in the optical regime are strongly modified, and so will be the resonant dipole-dipole interaction. Inside a band gap it is negligible [136, 137] and so is superradiance.

For free space, only the static dipole-dipole interaction is important at distances smaller than  $0.1\lambda$ . The distance-dependence of the SDDI inside an optical micro-cavity was recently measured in [138]. Both for free space and for the homogeneous dielectric of section 5.5.2, the static interaction varies as  $R^{-3}$ . This behavior is determined by optical modes with large wave vectors. The assumption that the dielectric is nondispersive entails that  $kc = n\omega_{\mathbf{k}}$  also for these large wave vectors and corresponding high frequencies. In reality,  $n(\omega)$  approaches 1 in the high-frequency limit. Thus, the dipole-dipole interaction at very short distances will differ less from the free-space case than was presented in figure 5.1. The same reasoning holds true for any dielectric: dispersion is important for static interactions at short distances.

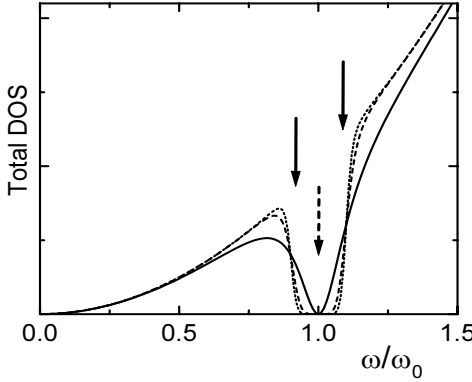
The static dipole-dipole interaction will not be modified strongly in a photonic crystal. In [21, 25] it was even assumed that the short-distance static interaction in a photonic crystal is the same as in free space because of frequency dispersion. More realistic values can be obtained by taking dispersion into account explicitly [139, 140]. Radiative shifts (including Lamb shifts) will also be influenced by frequency dispersion, for the same reason as given above for the static dipole-dipole interaction. In calculations of Lamb shifts in photonic crystals [20, 30, 141, 142], frequency dispersion is usually not taken into account. This makes the values obtained for the Lamb shift less accurate [142].

In [23], the interesting phenomenon of localization of superradiance at the edge of a photonic band gap showed up in calculations within the so-called “isotropic model”. In this model, the photonic crystal is assumed to have an isotropic dispersion relation and a diverging density of states at the band edges. It is known that the model is not very realistic and that band-edge effects strongly depend on the assumed photonic density of states [29, 30]. Here it is important to notice that the isotropic model assumes that all positions in the photonic crystal are equivalent: medium-induced detuning is absent. It will be interesting to study modified resonant dipole-dipole interactions in models which do include detuning. A summary of the remarks about modified superradiance in photonic crystals is given in figure 5.3.

## 5.8 Conclusions and outlook

In this chapter, it was studied how guest atoms are influenced by a photonic environment and by each other. This was done in a quantum mechanical model by integrating out the atomic dynamics and solving the Lippmann-Schwinger equation (5.12) for the Field operator of the total system. In chapter 4 it was derived that the atoms interact with a Field that is the sum of the electric field and their own polarisation fields. By taking into account this self-interaction, it was found in this chapter that the volume-integrated dynamical dipole field (5.28) of an atom is equal to minus one third of the polarisation field of that atom. This is an operator relation at finite frequency. Neither in a quantum nor in a classical theory is it necessary to add terms by hand to obtain the relation. The quantum and classical derivations follow a different route to the same result.

The spontaneous-emission rate of a single atom in a dielectric depends on the local density of states of the medium at the position of the dielectric. In section 5.3, emission rates and line shifts are derived from a T-matrix formalism. An empty-cavity local-field

**Figure 5.3:**

Sketch of three forms of total densities of states with a full photonic band gap. A quadratic dependence of the DOS on frequency (as known from homogeneous dielectrics) is assumed, interrupted by a gap. Well inside the gap (dashed arrow), spontaneous decay, resonant dipole-dipole interaction and superradiance are absent. Strongly modified superradiance is to be expected for frequencies at the edges of the band gap (solid arrows). Steeper band edges in the DOS will give stronger modifications of the dipole-dipole interaction. For interatomic distances much smaller than  $\lambda$ , the static dipole-dipole interaction dominates, which depends less strongly on the band gap. Frequency dispersion will be important for the static interaction at short distances.

factor was also (re-)derived for guest atoms in a homogeneous dielectric. The T-matrix formalism was then generalized in section 5.4 for an arbitrary finite number of guest atoms in the dielectric. The total Field operator is the sum of an atomic source-Field operator and an operator for the scattered Field. The source-field operator was used to calculate spontaneous-emission rates and line shifts. In the future, the formalism can be used to study multiple-scattered light and the survival of quantum properties of the initial quantum state of light in the scattered output.

An inhomogeneous medium modifies both the static and the resonant dipole-dipole interactions between atoms. In the multiple-scattering formalism of this chapter, both interactions have been identified in terms of the optical modes of the inhomogeneous medium. A static potential between the atoms is absent in the dipole Hamiltonian (5.4d) that was the basis of the calculations. The static and resonant dipole-dipole interactions appear “dynamically”, in the Lippmann-Schwinger equation for the two atoms [in Eq. (5.21)]. Both the static and the resonant dipole-dipole interactions are given in Eq. (5.71) in terms of the generalized-transverse true modes of the dielectric.

The multiple-scattering formalism has been used to study superradiance in a homogeneous medium. Modifications compared to the free-space case depend strongly on the interatomic distance. The present formalism can be directly used to study dipole-dipole interactions and superradiance in other specific dielectric geometries, for example in a Bragg mirror or in an “optical corral” [74, 76].

It is known that inhomogeneous line broadening is detrimental for cooperative effects [45]. The electronic component to inhomogeneous broadening will often be dominant for guest atoms in a solid environment, but electronic effects were neglected in this

chapter in order to focus on photonic effects. An important message from the calculations in this chapter is that differences in local line shifts and in local emission rates will also reduce superradiance. In photonic crystals, cavities and other dielectric structures, the local densities of states can vary strongly with position and frequency. Unlike the electronic detuning, the photonic component can not be modelled as a random variable: the LDOS is an engineered quantity that varies at a length scale much larger than an atom, yet smaller than an optical wavelength.

## Chapter 6

# Transient QED effects in absorbing dielectrics

*The spontaneous-emission rate of a radiating atom reaches its time-independent equilibrium value after an initial transient regime. The associated relaxation effects of the spontaneous-decay rate of atoms in dispersive and absorbing dielectric media are considered, for atomic transition frequencies near material resonances. A quantum mechanical description of such media is furnished by a damped-polariton model, in which absorption is taken into account through coupling to a bath. It is shown how all field and matter operators in this theory can be expressed in terms of the bath operators at an initial time. The consistency of these solutions for the field and matter operators are found to depend on the validity of certain velocity sum rules. The transient effects in the spontaneous-decay rate are studied with the help of several specific models for the dielectric constant, which are shown to follow from the general theory by adopting particular forms of the bath coupling constant.*

### 6.1 Introduction

In the previous chapters, frequency-dispersion and absorption of light in dielectrics were neglected. For example, in the chapters 2 and 3, the optical potentials of the plane scatterers were assumed real, so that their T-matrices satisfy an optical theorem and no light is lost in the planes. In the chapters 4 and 5, the dielectric function  $\epsilon(\mathbf{r})$  was assumed inhomogeneous but real and nondispersive. There is no principal difference between the electromagnetic field in such inhomogeneous dielectrics and in free space, in the sense that in both cases the field is a sum (or integral) over individual optical modes. The topic of this chapter is the quantum optical description of homogeneous dielectrics, which are described by complex and frequency-dependent dielectric functions  $\epsilon(\omega)$ . These dielectric functions satisfy the Kramers-Kronig relations [50, 51, 143, 144]. A description in terms of modes, each corresponding to a single frequency, will no longer be possible.

In quantum optics of linear dielectrics, one tries to describe the material medium in an

effective way with the help of the classical dielectric function  $\varepsilon(\mathbf{r}, \omega)$ , which, in general, is a complex function of both position and frequency and in this full generality describes the propagation and loss of light at each point in the dielectric. Sometimes it is possible to neglect the spatial variations, dispersion and losses altogether. The spontaneous-emission rate of an atom in such a simple dielectric is the refractive index  $n$  of the medium times the rate  $\Gamma_0$  in vacuum [117, 145–148], at least if local-field effects are neglected.

The situation becomes more complicated when material dispersion has to be taken into account [149–155]. Since the Kramers-Kronig relations tell that dispersion and loss always come together (be it not always at the same frequencies), one should like to include losses as well in order to describe all frequencies in one theory. The damped-polariton model [156–159] provides us with such a microscopic theory. From this theory it was shown that the radiative spontaneous-emission rate equals  $\Gamma_0$  times the real part of the refractive index at the transition frequency [160].

The quantum mechanical treatment of dissipative systems is more complicated than the classical one, because of the extra requirement that equal-time commutation relations do not change over time [45, 161]. Based on the damped-polariton model and on the fluctuation-dissipation theorem, phenomenological quantization theories were constructed that meet these requirements. In these theories, the dielectric function is an input function and the Maxwell field operators satisfy quantum Langevin equations with both loss and quantum noise terms [162, 163]. With the use of a Green-function approach, the phenomenological quantization theories have been generalized to inhomogeneous dielectrics, first for multilayer systems [164, 165] and later for general  $\varepsilon(\mathbf{r}, \omega)$  [166–170]. Field commutation relations turn out only to depend on the analytical properties of the Green function. However, the calculation of spontaneous emission inside such a medium would involve the actual computation of the Green function, which for general  $\varepsilon(\mathbf{r}, \omega)$  is not easy.

Interesting new effects have been predicted for photonic band-gap systems, such as photon-atom bound states and non-exponential spontaneous decay at the edges of the gap [21, 22, 171]. A current debate is whether the Weisskopf-Wigner approximation can be used in the calculation of spontaneous emission near an edge of a photonic band gap. This question seems to depend strongly on the analytic or singular behavior of the density of states at the edges of the gap, which has recently been calculated for face-centered-cubic and diamondlike crystal structures [29]. If near the edge of the band gap a large part of the modes has a cavitylike structure, producing nonzero dwell times near the emitting atom, then an emitted photon has a nonzero probability of being reabsorbed, which would give Rabi-like oscillations of the atomic population that are missed in the Weisskopf-Wigner approximation.

Nonexponential decay can also be caused by the interference of possible decay channels: for short times after the excitation of the atom, a larger frequency interval of the medium states plays a part in the decay process than for later times. Ultimately, only the refractive index at the atomic transition frequency plays a role, all in concordance with the energy-time uncertainty relation. This interference process already happens for spontaneous emission in vacuum. However, when the medium has a strong jump in the density of states around the atomic transition frequency, the interference effect will change substantially.

To separate the latter cause of nonexponential decay from the former, it is interesting to consider the spontaneous emission inside homogeneous lossy dielectrics with strong and narrow material resonances, where the density of states can also change very rapidly. Here real reabsorption processes do not play a role. In this chapter, we use the damped-polariton model formulated by Huttner and Barnett [157, 158] to study the interference effects of spontaneous emission. If absorption is neglected in the damped-polariton model, then we are left with the Hopfield model of a dielectric [152, 172], which has a frequency band gap inside which the refractive index is purely imaginary. The analogy between this polariton band gap system and photonic crystals was drawn in [173].

The organization of the chapter is as follows: in section 6.2 we introduce the theory and solve its equations of motion using Laplace transformations. In section 6.3 we show that the consistency of our solutions depends on the validity of a number of velocity sum rules, which are then proved. In section 6.4, we find that for long times all field operators can be expressed in terms of the initial bath operators, and we give an interpretation of the result. We also show how to relate the result to phenomenological quantization theories. Before we can discuss transient effects of spontaneous emission in section 6.6, we discuss in section 6.5 the Lorentz oscillator model and the point-scattering model. We show how both these models can be found from the damped-polariton theory by choosing a suitable coupling to the bath. The chapter ends with a discussion of the results and with conclusions in section 6.7.

## 6.2 The model and solutions of the equations of motion

The damped-polariton theory describes the interaction of light with an absorbing homogeneous medium. The coupling of the matter to a frequency continuum is the cause of the light absorption. The continuum could be a phonon bath or something else, but for the moment that is not specified: it is a collection of harmonic oscillators with a frequency-dependent coupling to the matter fields. Since the medium is homogeneous, the dynamics can be separated into a transverse and a longitudinal part. In this chapter we concentrate on the transverse excitations as described by the following Hamiltonian [157, 158]:

$$H = \int d^3k \left( \mathcal{H}_{\text{em}} + \mathcal{H}_{\text{mat}} + \mathcal{H}_{\text{bath}} + \mathcal{H}_{\text{int}} \right), \quad (6.1)$$

with

$$\mathcal{H}_{\text{em}} = \hbar \tilde{k} c a^\dagger(\lambda, \mathbf{k}) a(\lambda, \mathbf{k}), \quad (6.2a)$$

$$\mathcal{H}_{\text{mat}} = \hbar \tilde{\omega}_0 b^\dagger(\lambda, \mathbf{k}) b(\lambda, \mathbf{k}), \quad (6.2b)$$

$$\mathcal{H}_{\text{bath}} = \int_0^\infty d\omega \hbar \omega b_\omega^\dagger(\lambda, \mathbf{k}) b_\omega(\lambda, \mathbf{k}), \quad (6.2c)$$

$$\begin{aligned} \mathcal{H}_{\text{int}} &= \frac{1}{2} \int_0^\infty d\omega \hbar V(\omega) [b(\lambda, \mathbf{k}) + b^\dagger(\lambda, -\mathbf{k})] [b_\omega^\dagger(\lambda, \mathbf{k}) + b_\omega(\lambda, -\mathbf{k})] \\ &+ \frac{i}{2} \hbar \omega_c \sqrt{\frac{\tilde{\omega}_0}{\tilde{k} c}} [a(\lambda, \mathbf{k}) + a^\dagger(\lambda, -\mathbf{k})] [b^\dagger(\lambda, \mathbf{k}) - b(\lambda, -\mathbf{k})]. \end{aligned} \quad (6.2d)$$

We use the same notations as in [158]. In particular,  $\tilde{k}c$  stands for  $\sqrt{k^2 c^2 + \omega_c^2}$ , where the frequency  $\omega_c$  equals  $\alpha/\sqrt{\rho\varepsilon_0}$ , with  $\alpha$  the coupling constant between field and matter, and  $\rho$  the density. The resonance frequency  $\omega_0$  of the polarization field is renormalized to  $\tilde{\omega}_0$ , which is the positive-frequency solution of

$$\tilde{\omega}_0^2 = \omega_0^2 + \tilde{\omega}_0 \int_0^\infty d\omega V^2(\omega)/\omega. \quad (6.3)$$

The  $k$  integrals in the Hamiltonian are understood to also denote a summation over the two transverse polarization directions labeled by  $\lambda$ . The creation and annihilation operators satisfy standard bosonic commutation relations. The Heisenberg equations of motion for the bath annihilation operators are

$$\dot{b}_\omega(\lambda, \mathbf{k}, t) = -\frac{i}{2} V(\omega) [b(\lambda, \mathbf{k}, t) + b^\dagger(\lambda, -\mathbf{k}, t)] - i\omega b_\omega(\lambda, \mathbf{k}, t), \quad (6.4)$$

and similarly for the creation operators. In the following, we drop the  $(\lambda, \mathbf{k})$  labels. We solve implicitly for the bath variables as was done in [174] in a classical treatment of the model,

$$b_\omega(t) = -\frac{i}{2} V(\omega) \int_0^t dt' [b(t') + b^\dagger(t')] e^{-i\omega(t-t')} + b_\omega(0) e^{-i\omega t}. \quad (6.5)$$

The annihilation operators are defined in terms of the (transverse) physical fields

$$\begin{aligned} a(t) &= \sqrt{\frac{\varepsilon_0}{2\hbar\tilde{k}c}} [\tilde{k}cA(t) - iE(t)], \\ b(t) &= \sqrt{\frac{\rho}{2\hbar\tilde{\omega}_0}} [\tilde{\omega}_0X(t) + \frac{i}{\rho}P(t)], \end{aligned} \quad (6.6)$$

and similarly for the creation operators. Here  $A$  and  $E$  are the vector potential and the electric field, respectively,  $X$  the polarization field and  $P$  its canonical conjugate. Insertion of the solution (6.5) and its Hermitian conjugate in the equations of motion gives

$$\begin{aligned} \dot{E}(t) &= (\tilde{k}c)^2 A(t) + (\omega_c^2/\alpha)P(t), \\ \dot{A}(t) &= -E(t), \\ \dot{X}(t) &= (\omega_c/\alpha)^2 \varepsilon_0 P(t) + (\omega_c^2/\alpha)\varepsilon_0 A(t), \\ \dot{P}(t) &= -\alpha^2 \tilde{\omega}_0^2 / (\varepsilon_0 \omega_c^2) X(t) + \alpha^2 \tilde{\omega}_0 / (2\varepsilon_0 \omega_c^2) \int_0^t dt' F(t-t')X(t') - B(t). \end{aligned} \quad (6.7)$$

In the last equation, the bath operator  $B(t)$  is defined as

$$B(t) \equiv \sqrt{\frac{\hbar\tilde{\omega}_0\rho}{2}} \int_0^\infty d\omega_1 V(\omega_1) [b_{\omega_1}(0) e^{-i\omega_1 t} + b_{\omega_1}^\dagger(0) e^{i\omega_1 t}], \quad (6.8)$$

whereas the the function  $F$  in the convolution in (6.7) is:

$$F(t) \equiv 2 \int_0^\infty d\omega_1 V^2(\omega_1) \sin(\omega_1 t). \quad (6.9)$$

We get a system of algebraic equations by taking the Laplace transform, which we denote by a bar

$$\begin{pmatrix} p & -\tilde{k}^2 c^2 & 0 & -\omega_c^2/\alpha \\ 1 & p & 0 & 0 \\ 0 & -\varepsilon_0 \omega_c^2/\alpha & p & -\varepsilon_0 \omega_c^2/\alpha^2 \\ 0 & 0 & \bar{Q}(p) & p \end{pmatrix} \begin{pmatrix} \bar{E}(p) \\ \bar{A}(p) \\ \bar{X}(p) \\ \bar{P}(p) \end{pmatrix} = \begin{pmatrix} E(0) \\ A(0) \\ X(0) \\ P(0) - \bar{B}(p) \end{pmatrix}. \quad (6.10)$$

Here,  $\bar{Q}(p)$  is an abbreviation for  $\alpha^2 \tilde{\omega}_0^2 / (\varepsilon_0 \omega_c^2) [1 - \bar{F}(p)/(2\tilde{\omega}_0)]$ . Through the operator  $\bar{B}(p)$  the bath remains part of the system of equations: this is as far as we can “integrate out” the bath variables.

Now we can determine the dielectric function  $\varepsilon(\omega)$ , which is a *classical* quantity, by putting the determinant of the  $(4 \times 4)$  coefficient matrix to zero. The determinant gives the dispersion relation

$$\bar{D}(p) \equiv \bar{\varepsilon}(p)p^2 + k^2 c^2 = 0, \quad (6.11)$$

with the “Laplace dielectric function”

$$\bar{\varepsilon}(p) = 1 + \frac{\omega_c^2}{p^2 + \tilde{\omega}_0^2 - \frac{1}{2}\tilde{\omega}_0 \bar{F}(p)}. \quad (6.12)$$

The function  $\bar{F}(p)$  is the Laplace transform of  $F(t)$ , which was defined in Eq. (6.9). From this we find the dielectric function

$$\varepsilon(\omega) = \bar{\varepsilon}(-i\omega + \eta) = 1 - \frac{\omega_c^2}{\omega^2 - \tilde{\omega}_0^2 + \frac{1}{2}\tilde{\omega}_0 F(\omega)}, \quad (6.13)$$

with infinitesimal positive  $\eta$  and

$$F(\omega) \equiv \bar{F}(-i\omega + \eta) = \int_0^\infty d\omega_1 V^2(\omega_1) \left( \frac{1}{\omega_1 - \omega - i\eta} + \frac{1}{\omega_1 + \omega + i\eta} \right). \quad (6.14)$$

The difference between  $F(\omega)$  and  $F(t)$  is denoted by their arguments. The dielectric function satisfies the Kramers-Kronig relations and has the property of a response function that  $\varepsilon(-\omega^*)$  equals  $\varepsilon^*(\omega)$ . It can be shown that it has no poles in the upper half plane provided that the integral in Eq. (6.3) exists. Previous authors [158, 174–176] assumed that the analytical continuation of  $V^2(\omega)$  to negative frequencies is antisymmetrical in frequency. Then Eq. (6.13) reduces to the dielectric constant in [175] where it was shown to be identical to the more complicated expression in [158].

We combine Eqs. (6.10) and (6.12) and write the Laplace fields in terms of the fields at time  $t = 0$  with coefficients that are functions of the Laplace dielectric function  $\bar{\varepsilon}(p)$  and susceptibility  $\bar{\chi}(p) = \bar{\varepsilon}(p) - 1$ . For the electric field we find

$$\begin{aligned} \bar{E}(p) &= \bar{D}^{-1}(p) \left\{ pE(0) + [p^2 \bar{\chi}(p) + k^2 c^2] A(0) \right. \\ &\quad \left. + \frac{\alpha}{\varepsilon_0} p \left[ \frac{p^2}{\omega_c^2} \bar{\chi}(p) - 1 \right] X(0) + \frac{1}{\alpha} p^2 \bar{\chi}(p) [P(0) - \bar{B}(p)] \right\}. \end{aligned} \quad (6.15)$$

The other Laplace operators can be found in the same way and are listed in appendix D. The inverse Laplace transform gives the fields at time  $t$  in terms of the fields at time  $t = 0$ ,

$$E(t) = M_{EE}(t)E(0) + M_{EA}(t)A(0) + M_{EX}(t)X(0) + M_{EP}(t)P(0) + B_E(t), \quad (6.16)$$

where, for instance,

$$M_{EE}(t) = \frac{1}{2\pi i} \int_{-i\infty}^{i\infty} dp e^{pt} \bar{D}^{-1}(p) p. \quad (6.17)$$

The operator  $B_E(t)$  in Eq. (6.16) is the contribution of the  $t = 0$  bath operators to the electric field. This term will be analyzed in more detail in section 6.4.

The *equal*-time commutation relations of the field operators are

$$[A(\lambda, \mathbf{k}, t), -\varepsilon_0 E(\lambda', -\mathbf{k}', t)] = [X(\lambda, \mathbf{k}, t), P(\lambda', -\mathbf{k}', t)] = i\hbar \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}'). \quad (6.18)$$

All other inequivalent combinations of operators commute. In particular  $A$  and  $X$  are independent canonical variables. Hence, we have the property  $[A, -D] = [A, -\varepsilon_0 E]$  with the displacement field  $D$  defined as  $\varepsilon_0 E - \alpha X$ . With the help of Eqs. (6.16) and (6.18), we can also calculate nonequal-time commutators, for example:

$$\begin{aligned} [E(\lambda, \mathbf{k}, t), E(\lambda', -\mathbf{k}', 0)] &= M_{EA}(t) [A(\lambda, \mathbf{k}, 0), E(\lambda', -\mathbf{k}', 0)] \\ &= -\frac{i\hbar}{\varepsilon_0} M_{EA}(t) \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}'). \end{aligned} \quad (6.19)$$

In principle we have solved the complete time evolution of the field operators. In section 6.3 we analyze in more detail their short-time behavior, whereas in section 6.4 we consider the long-time limit.

### 6.3 Short-time limit: sum rules

For fixed  $k$ , the zeroes of the dispersion relation (6.11) are the poles of the integrand in Eq. (6.17). We assume that they are simple first-order poles and rewrite the integral (6.17) as an integral over frequencies  $\omega = ip$ . Then, using contour integration in the lower frequency half plane, we find the coefficients for the electric field in Eq. (6.16) as

$$\begin{aligned} M_{EE}(t) &= \sum_j \text{Re} \left[ \frac{v_{pj} v_{gj}}{c^2} e^{-i\Omega_j t} \right], \\ M_{EA}(t) &= -kc \sum_j \text{Im} \left[ \frac{v_{pj}^2 v_{gj}}{c^3} e^{-i\Omega_j t} \right], \\ M_{EX}(t) &= -\frac{\alpha k^2 c^2}{\omega_c^2 \varepsilon_0} \sum_j \text{Re} \left[ \frac{v_{pj} v_{gj}}{c^2} \left( 1 - \frac{v_{pj}^2}{c^2} + \frac{\omega_c^2}{k^2 c^2} \right) e^{-i\Omega_j t} \right], \\ M_{EP}(t) &= \frac{kc}{\alpha} \sum_j \text{Im} \left[ \frac{v_{gj}}{c} \left( 1 - \frac{v_{pj}^2}{c^2} \right) e^{-i\Omega_j t} \right]. \end{aligned} \quad (6.20)$$

Some details of the calculation and a list of coefficients  $M_{mn}(t)$  of other operators can be found in appendix D. In these expressions, the frequencies  $\Omega_j = \Omega_j(k)$  are the complex-frequency solutions of the dispersion relation  $\omega^2 \varepsilon(\omega) - k^2 c^2 = 0$ . All  $\Omega_j(k)$  have a negative imaginary part. Since  $\varepsilon(-\omega^*) = \varepsilon^*(\omega)$ , it follows that  $-\Omega_j^*(k)$  is also a solution of the dispersion relation. We can choose  $\Omega_j(k)$  to be the solution with a positive real part. The summation over  $j$  is a summation over all the polariton branches of the medium. For each branch, the complex phase velocity is defined as  $v_{pj}(k) = \Omega_j(k)/k$  and the group velocity as  $v_{gj}(k) = d\Omega_j(k)/dk$ . For convenience, we leave out their explicit  $k$  dependence in the following.

From equation (6.16) we can see that the “diagonal” coefficient  $M_{EE}(t)$  in Eq. (6.20) should have the value 1 at time  $t = 0$  and the “off-diagonal” coefficients  $M_{EA}(0)$ ,  $M_{EX}(0)$ , etc. should have the value 0. The coefficients of the other field operators should also follow this rule. If these constraints are satisfied, the nonequal-time commutators like Eq. (6.19) get the right equal-time limits as well. The coefficients [Eq. (6.20)] can only have the right  $t = 0$  limits if certain velocity sum rules are satisfied.

Velocity sum rules can be derived in a systematic way by evaluating the following two types of integrals:

$$\int_{-\infty}^{\infty} d\omega \frac{(\omega + i\delta)^n}{\varepsilon(\omega)\omega^2 - k^2 c^2}, \quad \text{for } n = -1, 0, 1, \quad (6.21a)$$

$$\int_{-\infty}^{\infty} d\omega \frac{(\omega + i\delta)^m \chi(\omega)}{\varepsilon(\omega)\omega^2 - k^2 c^2}, \quad \text{for } m = -1, 0, 1, 2, 3. \quad (6.21b)$$

Here  $\varepsilon(\omega)$  is an *arbitrary* dielectric function that satisfies the Kramers-Kronig relations, so it is not necessarily of the specific form (6.13). The integrals can be evaluated using contour integration in the complex-frequency plane. We can close the contours either in the upper or in the lower half plane. Equating the two answers gives a velocity sum rule. In this way one finds for all wavevectors  $k$

$$\sum_j \text{Re}(v_{gj} v_{pj} / c^2) = 1, \quad (6.22)$$

$$\sum_j \text{Re}(v_{gj} / v_{pj}) = 1. \quad (6.23)$$

These sum rules can be found from Eq. (6.21a) with  $n = 1$  and  $n = -1$ , respectively. Both relations have been obtained before [152, 158, 177, 178]. The second was coined the Huttner-Barnett sum rule in [177] because of its importance in phenomenological quantum theories of dielectrics. A second group of sum rules has the form

$$\sum_j \text{Im}(v_{gj} v_{pj}^{2q}) = 0, \quad \text{for } q = -1, 0, 1. \quad (6.24)$$

The rules with  $q = -1$  and  $q = 1$  follow from Eq. (6.21b) with  $m = 0$  and  $m = 2$ , respectively; the case with  $q = 0$  follows from Eq. (6.21a) with  $n = 0$ .

All of these sum rules are independent of any specific form of the dielectric function as long as it satisfies the Kramers-Kronig relations. Other sum rules do depend on the

behavior of  $\varepsilon(\omega)$  for high or low frequencies. For example, from Eq. (6.21b) with  $m = -1$  we find

$$\sum_j \operatorname{Re}(c^2 v_{gj}^3 / v_{pj}^3) = \varepsilon(0). \quad (6.25)$$

This sum rule depends on the static limit of the dielectric function. For conductors the dielectric function is singular at  $\omega = 0$  [4], but for dielectric functions that can be found from the damped-polariton model,  $\varepsilon(0)$  is finite. Two other sum rules can be derived when for high frequencies  $\omega^2 \chi(\omega)$  approaches a constant value that we name  $-\omega_{\text{lim}}^2$ . From Eq. (6.21b) with  $m = 3$ , we then find

$$\sum_j \operatorname{Re}(v_{gj} v_{pj}^3 / c^4) = 1 + \omega_{\text{lim}}^2 / (kc)^2. \quad (6.26)$$

Moreover, if  $\omega^2 \chi(\omega) + \omega_{\text{lim}}^2$  falls off faster than  $\omega^{-1}$ , then the integral

$$\int_{-\infty}^{\infty} d\omega \frac{\omega^2 [\omega^2 \chi(\omega) + \omega_{\text{lim}}^2]}{\varepsilon(\omega) \omega^2 - k^2 c^2} \quad (6.27)$$

produces the sum rule (6.24) with  $q = 2$ .

Returning now to the time-dependent coefficients (6.20) (and the other ones in appendix D), one finds, by inspection, that one needs all the above sum rules except Eq. (6.25) to prove that the coefficients have the right limits for  $t = 0$ . In particular, from Eq. (6.13) it follows that the frequency  $\omega_{\text{lim}}$  as defined above exists in the damped-polariton model and equals  $\omega_c$ . Then with Eqs. (6.23) and (6.26) we see that indeed one has  $M_{EX}(0) = 0$  in Eq. (6.20).

It is easy to prove the above sum rules in the following one-resonance model:

$$\varepsilon(\omega) = 1 - \frac{\omega_c^2}{\omega^2 - \omega_0^2}. \quad (6.28)$$

This  $\varepsilon(\omega)$  is real and violates the Kramers-Kronig relations, but it can be considered as a limiting case of an acceptable dielectric function. The high-frequency limit of  $\omega^2 \chi(\omega)$  indeed equals  $-\omega_c^2$ . The two sum rules (6.22), (6.23) were shown to be valid for this model [158] and we want to check Eq. (6.26) as well. The dispersion relation is

$$\omega^4 - (\omega_0^2 + \omega_c^2 + k^2 c^2) \omega^2 + k^2 c^2 \omega_0^2 = 0, \quad (6.29)$$

which has two (real) solutions  $\Omega_+^2$  and  $\Omega_-^2$  with sum  $(\omega_0^2 + \omega_c^2 + k^2 c^2)$  and product  $k^2 c^2 \omega_0^2$ . It follows that for all  $k$

$$\begin{aligned} v_{p+}^3 v_{g+} + v_{p-}^3 v_{g-} &= \frac{1}{4k^3} \frac{d}{dk} (\Omega_+^4 + \Omega_-^4) \\ &= \frac{1}{4k^3} \frac{d}{dk} [(\Omega_+^2 + \Omega_-^2)^2 - 2\Omega_+^2 \Omega_-^2] = \left(1 + \frac{\omega_c^2}{k^2 c^2}\right) c^4, \end{aligned} \quad (6.30)$$

in agreement with Eq. (6.26). The other sum rules can also be checked for this simple model. The sum rules [Eq. (6.24)] obviously hold, because all group and phase velocities are real in this model. In models that respect the Kramers-Kronig relations, these sum rules are nontrivial.

## 6.4 Long-time limit

### 6.4.1 Field and medium operators

The coefficients  $M_{EE}(t)$ , etc., in Eq. (6.20) damp out exponentially in time. Every polariton branch has its own characteristic damping time  $\tau_j(k) = 1/[\text{Im } \Omega_j(k)]$ . After a few times the maximum characteristic damping period with the maximum taken over all branches, the exponentially damped coefficients can be neglected. We call this the long-time limit. The speed at which it is attained depends on  $\varepsilon(\omega)$  and on  $k$ . For long times, only the bath operator  $B_E(t)$  in Eq. (6.16) survives, because it has poles on the imaginary axis in the complex  $p$  plane:

$$B_E(t) = -\frac{1}{2\pi i \omega_c} \sqrt{\frac{\hbar \tilde{\omega}_0}{2\varepsilon_0}} \int_0^\infty d\omega_1 V(\omega_1) \times \\ \times \int_{-i\infty}^{i\infty} dp e^{pt} \frac{p^2 \bar{\chi}(p)}{\bar{\varepsilon}(p)p^2 + k^2 c^2} \left[ \frac{b_{\omega_1}(0)}{p + i\omega_1} + \frac{b_{\omega_1}^\dagger(0)}{p - i\omega_1} \right]. \quad (6.31)$$

Hence, in the long-time limit, all field operators are functions of the initial bath operators alone. For the electric field we find that  $E(t)$  tends to  $E_l(t)$ , with

$$E_l(t) = \int_0^\infty d\omega_1 [E_l^+(\omega_1)e^{-i\omega_1 t} + \text{H.c.}] \quad (6.32)$$

where the subscript  $l$  denotes the long-time limit; “H.c.” means Hermitian conjugate. The temporal (and spatial) Fourier components of the long-time solutions of the electric and other fields are

$$E_l^+(\omega) = -\frac{1}{\omega_c} \sqrt{\frac{\hbar \tilde{\omega}_0}{2\varepsilon_0}} \frac{V(\omega) \omega^2 \chi(\omega) b_\omega(0)}{\varepsilon(\omega) \omega^2 - k^2 c^2}, \\ A_l^+(\omega) = \frac{i}{\omega_c} \sqrt{\frac{\hbar \tilde{\omega}_0}{2\varepsilon_0}} \frac{V(\omega) \omega \chi(\omega) b_\omega(0)}{\varepsilon(\omega) \omega^2 - k^2 c^2}, \\ X_l^+(\omega) = -\frac{1}{\alpha \omega_c} \sqrt{\frac{\hbar \tilde{\omega}_0 \varepsilon_0}{2}} \frac{V(\omega) (\omega^2 - k^2 c^2) \chi(\omega) b_\omega(0)}{\varepsilon(\omega) \omega^2 - k^2 c^2}, \\ P_l^+(\omega) = \frac{i\alpha}{\omega_c^3} \sqrt{\frac{\hbar \tilde{\omega}_0}{2\varepsilon_0}} \frac{V(\omega) (\omega^2 - k^2 c^2 - \omega_c^2) \omega \chi(\omega) b_\omega(0)}{\varepsilon(\omega) \omega^2 - k^2 c^2}, \quad (6.33)$$

where the superscript  $+$  denotes the positive-frequency component of the operator. For future reference we also give the long-time limit of the electric field operator as a function of position and time

$$\mathbf{E}_l(\mathbf{r}, t) = -\sqrt{\frac{\hbar \tilde{\omega}_0}{2(2\pi)^3 \varepsilon_0 \omega_c^2}} \int d\mathbf{k} \sum_{\lambda=1,2} \mathbf{e}_\lambda(\mathbf{k}) \times \\ \times \int_0^\infty d\omega_1 \left[ \frac{V(\omega_1) \omega_1^2 \chi(\omega_1) b_{\omega_1}(\lambda, \mathbf{k}, 0)}{\varepsilon(\omega_1) \omega_1^2 - k^2 c^2} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_1 t)} + \text{H.c.} \right]. \quad (6.34)$$

Similar expressions can be given for the other operators. Notice that these long-time solutions indeed are solutions of the equations of motion (6.7) and of the Maxwell equations. The canonical commutation relations (6.18) should be preserved in this long-time limit. Also, the nonequal-time commutation relations like in eq (6.19) should be time-translation invariant. The commutation relations can be verified with the equality

$$\frac{\pi\tilde{\omega}_0}{2\omega_c^2} V^2(\omega) |\chi(\omega)|^2 = \text{Im } \chi(\omega) = \text{Im } \varepsilon(\omega) \equiv \varepsilon_i(\omega), \quad (6.35)$$

which follows from Eqs. (6.13) and (6.14). Since  $\varepsilon_i(\omega)$  is antisymmetric in  $\omega$ , all commutators can be shown to be proportional to integrals over the whole real-frequency axis. Contour integration then leads to the required results.

The solutions found above can be related to those obtained by explicit diagonalization of the full Hamiltonian of the model. In [158], this diagonalization was carried out by using Fano's technique. In that way the field and medium operators were written in terms of the diagonalizing annihilation operators [called  $C(\mathbf{k}, \omega)$  in [158]] and the corresponding creation operators. If one replaces the bath annihilation operators  $b_\omega(\mathbf{k}, 0)$  in the long-time solutions (6.33) by the diagonalizing annihilation operators  $C(\mathbf{k}, \omega)$  and if one makes similar replacements for the creation operators, the expressions for the field and medium operators in [158] are recovered.

The long-time solutions can be interpreted as follows: when the dielectric medium is prepared in a state that is not an eigenstate of the Hamiltonian and if the coupling  $V(\omega)$  is nonzero for all the frequencies, then the medium tends to an equilibrium that is determined by the state of the bath. The time taken for this equilibrium to settle down is the time after which the long-time solutions can be used for the field operators. So, one can always use the long-time solutions in the calculations unless the medium has been specially prepared in a nonequilibrium state a short time before one does the experiment. The interpretation of the long-time solution will become clearer in section 6.6 where we calculate spontaneous-emission rates.

In summary, for times long after  $t = 0$ , all field operators can be expressed solely in terms of the bath operators at time  $t = 0$ . The time evolution is governed by the bath Hamiltonian alone. The field operators still satisfy Maxwell's equations and the canonical commutation relations. Classical expressions for the Maxwell fields would have died exponentially to zero in this long-time limit.

## 6.4.2 Relation with phenomenological theories

The long-time solutions of the field operators can be related to expressions in phenomenological theories, as we will show presently. In phenomenological quantum mechanical theories of homogeneous absorbing dielectrics [162–165], a noise current density operator  $\mathbf{J}$  is added to the Maxwell equations in order to preserve the field commutation relations

$$\nabla \times \mathbf{E}^+(\mathbf{r}, \omega) = i\omega \mathbf{B}^+(\mathbf{r}, \omega), \quad (6.36a)$$

$$\nabla \times \mathbf{B}^+(\mathbf{r}, \omega) = -i\omega\mu_0 \tilde{\mathbf{D}}^+(\mathbf{r}, \omega) + \mu_0 \mathbf{J}^+(\mathbf{r}, \omega). \quad (6.36b)$$

The displacement field  $\tilde{\mathbf{D}}^+$  in the last equation is defined in terms of the electric field and the dielectric function as

$$\tilde{\mathbf{D}}^+(\mathbf{r}, \omega) = \varepsilon_0 \varepsilon(\omega) \mathbf{E}^+(\mathbf{r}, \omega). \quad (6.37)$$

We write  $\tilde{\mathbf{D}}$  to stress the difference with the microscopic displacement field  $\mathbf{D}$  in section 6.2. After taking the spatial Fourier transform and using  $\mathbf{B}^+ = \nabla \times \mathbf{A}^+$  and  $\mathbf{E}^+ = i\omega \mathbf{A}^+$ , so that the first of the Maxwell equations is satisfied, one finds from the second equation

$$\left[ \omega^2 \varepsilon(\omega) - k^2 c^2 \right] A^+(\lambda, \mathbf{k}, \omega) = -\frac{1}{\varepsilon_0} J^+(\lambda, \mathbf{k}, \omega). \quad (6.38)$$

The vector potential and all Maxwell fields can be calculated in terms of the noise current density  $J$ . The canonical commutation relations are preserved if for the noise current density one chooses [162, 163]

$$\left[ J^+(\lambda, \mathbf{k}, \omega), [J^+(\lambda', \mathbf{k}', \omega')]^\dagger \right] = \frac{\hbar \omega^2 \varepsilon_0 \varepsilon_i(\omega)}{\pi} \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (6.39)$$

Instead of using the noise current density, one defines basic bosonic operators

$$f(\lambda, \mathbf{k}, \omega) = \sqrt{\frac{\pi}{\hbar \omega^2 \varepsilon_0 \varepsilon_i(\omega)}} J^+(\lambda, \mathbf{k}, \omega), \quad (6.40)$$

so that these operators satisfy simple commutation relations

$$\left[ f(\lambda, \mathbf{k}, \omega), f^\dagger(\lambda', \mathbf{k}', \omega') \right] = \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}') \delta(\omega - \omega'). \quad (6.41)$$

Now we turn to the long-time solutions of the field operators that we determined in section 6.4.1. The long-time solution of the vector potential in Eq. (6.33) obviously is a solution of the following inhomogeneous wave equation:

$$\left[ \varepsilon(\omega) \omega^2 - k^2 c^2 \right] A_l^+(\lambda, \mathbf{k}, \omega) = \frac{i}{\omega_c} \sqrt{\frac{\hbar \tilde{\omega}_0}{2 \varepsilon_0}} V(\omega) \omega \chi(\omega) b_\omega(\lambda, \mathbf{k}, 0). \quad (6.42)$$

This kind of equation is well known in Langevin theories [45, 161]: the coupling to a bath gives a damping term (here: a complex dielectric constant) in the equations of motion of the system. Besides damping, there is an extra term that is neglected classically. This term is the quantum noise operator, which features the bath operators at time  $t = 0$ .

The long-time solution (6.42) can justify the phenomenological equation (6.38) if we identify

$$f_l(\lambda, \mathbf{k}, \omega) = -i \frac{V(\omega) \chi(\omega)}{|V(\omega) \chi(\omega)|} b_\omega(\lambda, \mathbf{k}, 0), \quad (6.43)$$

where we used Eq. (6.35). We see that up to a phase factor, the bath operators  $b_\omega(\lambda, \mathbf{k}, 0)$  from the microscopic theory serve as basic bosonic operators  $f(\lambda, \mathbf{k}, \omega)$  in the phenomenological theories. We want to stress that the identification (6.43) is only valid in the long-time limit when the medium is in equilibrium with the bath.

In section 6.2 we saw that  $-\varepsilon_0 E$  is the canonical conjugate field of  $A$  and that  $[A, -D]$  gives the canonical result as well. Since we can make the identification (6.43), the same relations hold in the phenomenological theory that was described in this section. But now let us calculate the commutator  $[A, -\tilde{D}]$  with  $\tilde{D}^+$  defined as in Eq. (6.37) and  $\tilde{D}^-$  as its Hermitian conjugate. We can use the long-time solutions because the commutation relations are preserved

$$[A(\lambda, \mathbf{k}, t), -\tilde{D}(\lambda', -\mathbf{k}', t)] = \frac{2i\hbar}{\pi} \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}') \int_0^\infty d\omega \frac{\varepsilon_r(\omega) \varepsilon_i(\omega) \omega^3}{|\varepsilon(\omega) \omega^2 - k^2 c^2|^2}, \quad (6.44)$$

with  $\varepsilon_r(\omega)$  the real part of the dielectric constant. The symmetry of the integrand enables us to rewrite the right-hand side as an integral over all real frequencies. When using contour integration, one cannot replace  $\varepsilon^*(\omega)$  by  $\varepsilon(-\omega^*)$ , but the analytical continuation to complex frequencies of  $\varepsilon^*(\omega) = \varepsilon(-\omega)$  must be used instead

$$[A(\lambda, \mathbf{k}, t), -\tilde{D}(\lambda', -\mathbf{k}', t)] = i\hbar \delta_{\lambda\lambda'} \delta(\mathbf{k} - \mathbf{k}') \sum_j \text{Re} [\varepsilon(-\Omega_j) v_{pj} v_{gj} / c^2], \quad (6.45)$$

where we assumed, as before, that all poles of the dispersion relation are first-order poles. Note that  $\varepsilon(-\Omega_j)$  depends on the behavior of the dielectric function in the *upper* half plane. Contrary to a statement in [163], the commutator does not give the canonical result because in general there is no sum rule for the right-hand side of the equation. In other words,  $(D - \varepsilon_0 E)$  is canonically independent of  $E$ , but  $(\tilde{D} - \varepsilon_0 E)$  is not. The operator  $(D - \tilde{D})$  is proportional to the Langevin noise term in the wave equation for the electric field.

Now let us neglect absorption at all frequencies. Strictly speaking, the limit  $\varepsilon_i(\omega) \rightarrow 0$  is unphysical because it violates the Kramers-Kronig relations, but the limit is sometimes taken for dielectrics that show negligible absorption at optical frequencies [164, 165, 177]. When  $\varepsilon(\omega)$  becomes real, the solutions  $\Omega_j$  become real and in that limit one has  $\varepsilon(-\Omega_j) \rightarrow \varepsilon(\Omega_j) = (c/v_{p,j})^2$ . Inserting this in Eq. (6.45) and using the Huttner-Barnett sum rule  $\sum_j \text{Re} (v_{gj}/v_{pj}) = 1$ , we immediately find the canonical result for  $[A, -\tilde{D}]$ . We compare this with the results in [177] where the dielectric function is assumed to be real. There a phenomenological Lagrangian was introduced and the fields  $A$  and  $-\tilde{D}$  were correctly identified as a canonical pair. The Huttner-Barnett sum rule was invoked to show that their commutator indeed had the canonical form. It was concluded that it is misleading that also  $[A, -\varepsilon_0 E]$  has the canonical form. Here we have learnt that this misleading result is not surprising: in the limit of real dielectric constants *and only then*, both  $[A, -\varepsilon_0 E]$  and  $[A, -\tilde{D}]$  can have the canonical form in the same gauge, the reason being that  $\tilde{D}$  approaches  $D$  in that limit.

## 6.5 Model dielectric functions

Phenomenological theories as discussed in section 6.4.2 have expressions for  $\varepsilon(\omega)$  as input. In practice, this input will be the outcome of measurements of the dielectric function. By choosing the appropriate microscopic coupling constants and resonance frequencies in

the damped-polariton model, one can hope to find a given dielectric function, thus providing a connection with phenomenological theories. It was argued in [174, 175] that the well-known Lorentz oscillator form of the dielectric function could not be found from the damped-polariton theory in this way. We shall reconsider this issue below.

A dielectric function that follows from the damped-polariton Hamiltonian (6.2a) will have a single resonance because there is only one resonance frequency  $\omega_0$  in the matter fields. Experimentally, one may find more resonances in  $\varepsilon(\omega)$ . This should not be used as an objection to the damped-polariton model because, in principle, one could easily extend the theory with more material resonances. In this section, we consider two of these one-resonance models.

### 6.5.1 The Lorentz oscillator model

We want to find microscopic coupling constants in the damped-polariton theory so that the resulting  $\varepsilon(\omega)$  has the following Lorentz oscillator form

$$\varepsilon_{\text{Lor}}(\omega) = 1 - \frac{\omega_{c,\text{Lor}}^2}{\omega^2 - \omega_{\text{res}}^2 + 2i\omega\kappa_0}. \quad (6.46)$$

Here  $\omega_{\text{res}}$  is the resonance frequency of the medium and  $\omega_{c,\text{Lor}}$  is a frequency that is related to the coupling strength between the electromagnetic and the matter field. Identifying  $\varepsilon(\omega)$  from Eq. (6.13) with  $\varepsilon_{\text{Lor}}(\omega)$ , we find, apart from the trivial identification  $\omega_c = \omega_{c,\text{Lor}}$ ,

$$\int_0^\infty d\omega_1 V^2(\omega_1) \left[ \frac{1}{\omega_1 - \omega - i\eta} + \frac{1}{\omega_1 + \omega + i\eta} \right] = 4 \left( \frac{\omega}{\tilde{\omega}_0} \right) i\kappa_0 + \Delta, \quad (6.47)$$

where the frequency shift  $\Delta$  is defined such that  $\omega_{\text{res}}^2 = \tilde{\omega}_0^2 - \tilde{\omega}_0\Delta/2$ . The coupling  $V^2(\omega_1)$  is fixed by the identification of the imaginary parts and for all frequencies it equals  $V^2(\omega_1) = 4\kappa_0\omega_1/(\pi\tilde{\omega}_0)$ . However, if we insert this coupling in the equation for the real parts, we find that the frequency shift  $\Delta$  is infinitely large. Also, the renormalized frequency  $\tilde{\omega}_0$  in Eq. (6.3) blows up. We can solve this problem by introducing a frequency cutoff in the coupling, namely,  $V^2(\omega_1) = 4\kappa(\omega_1)\omega_1/(\pi\tilde{\omega}_0)$  with

$$\kappa(\omega_1) = \frac{\kappa_0\Omega^2}{\Omega^2 + \omega_1^2}. \quad (6.48)$$

With this choice one finds  $\tilde{\omega}_0 = \sqrt{\omega_0^2 + 2\kappa_0\Omega}$ , which clearly has a strong dependence on the cutoff frequency. The shift  $\Delta$  becomes both finite and frequency-dependent

$$\Delta(\omega) = \frac{4}{\pi\tilde{\omega}_0} \mathcal{P} \int_0^\infty d\omega_1 \omega_1 \kappa(\omega_1) \left( \frac{1}{\omega_1 - \omega} + \frac{1}{\omega_1 + \omega} \right) = 4\kappa(\omega) \frac{\Omega}{\tilde{\omega}_0}. \quad (6.49)$$

The principal-value integral can be evaluated by means of contour integration in the complex-frequency plane. In this way we arrive at the following expression for the dielectric function

$$\varepsilon(\omega) = 1 - \frac{\omega_c^2}{\omega^2 - \omega_0^2 - 2\Omega[\kappa_0 - \kappa(\omega)] + 2i\omega\kappa(\omega)}. \quad (6.50)$$

We can choose  $\Omega$  arbitrarily high (but finite). Quite unlike  $\tilde{\omega}_0$ , the optical resonance frequency  $\omega_{\text{res}}$  approaches  $\omega_0$  from above, the higher we choose the cutoff, since one has  $\omega_{\text{res}}^2 \simeq \omega_0^2 + 2\kappa_0\omega_0^2/\Omega$ . Note that the dielectric function (6.50) has the right high-frequency limit  $\omega^2\chi(\omega) \rightarrow -\omega_c^2$  as required in section 6.3.

It is well known that there are two branches of solutions of the dispersion relation when the dielectric function is of the form (6.46): an upper and a lower polariton branch. The dielectric function (6.50) gives rise to *another* branch: it has a purely imaginary frequency with magnitude of the order of the cutoff frequency. This “cutoff branch” has negligible  $k$  dependence. In fact, the leading  $k$ -dependent term for large  $\Omega$  is  $2i\omega_c^2\kappa_0k^2c^2/\Omega^4$ . Clearly, the group velocity on this branch is practically zero so that the contribution of the cutoff branch to the velocity sum rules of section 6.3 can be neglected.

We conclude that high cutoff frequencies can be chosen such that in the optical frequency regime the dielectric function cannot be discerned from a Lorentz dielectric function with resonance frequency  $\omega_{\text{res}} = \omega_0$  and damping constant  $\kappa_0$ . The solutions of the dispersion relation of the upper and the lower polariton branch together satisfy the sum rules of section 6.3.

## 6.5.2 The point-scattering model

In general the dielectric function  $\varepsilon(\omega)$  describes the propagation of a coherent light beam in a fixed direction in an isotropic medium. A complex  $\varepsilon(\omega)$  means that there is extinction, which can be caused either by scattering or absorption or both. The dielectric function does not contain information about the extinction mechanism. A well-known dielectric medium showing polariton behavior is the dilute gas, which can be described as a collection of point dipoles that scatter light independently. If only one type of elastic scatterers is present, each having only one resonance, then the dielectric function is given by [68]

$$\varepsilon_{\text{sc}}(\omega) = 1 - \frac{4\pi c^2 \Gamma_e n}{\omega^2 - \omega_{\text{res}}^2 + \frac{2}{3}i\Gamma_e \omega^3/c}, \quad (6.51)$$

where  $n = N/V$  is the density of the scatterers [not to be confused with the refractive index  $n(\omega)$ ] and  $\Gamma_e = e^2/(4\pi\varepsilon_0 m_e c^2)$  is the classical electron radius. This dielectric function can also be found if one supposes that the medium consists of classical harmonically bound point charges whose motion is described by the Abraham-Lorentz equation. The dielectric function (6.51) has the property that the corresponding T-matrix  $t(\omega)$  satisfies the optical theorem with  $t(\omega)$  defined as  $\varepsilon(\omega) = 1 - nt(\omega)(c/\omega)^2$ . However, Eq.(6.51) is not a proper response function, since it has a pole near the very large *positive* imaginary frequency  $3ic/(2\Gamma_e)$ . This can be related to the need for the a-causal phenomenon called preacceleration to avoid so-called runaway solutions of the Abraham-Lorentz equation [179].

Although we know that in the damped-polariton theory only proper response functions can be found, we proceed like in the previous subsection and try to find coupling constants that in the optical regime give rise to the dielectric function (6.51). Equating

with Eq. (6.13) we get  $\omega_c^2 = 4\pi c^2 \Gamma_e n$  and  $V^2(\omega_1) = 4\Gamma(\omega_1)\omega_1^3/(3\pi\tilde{\omega}_0 c)$  with

$$\Gamma(\omega_1) = \frac{\Gamma_e \Omega^4}{\Omega^4 + \omega_1^4}. \quad (6.52)$$

Here we have inserted a convenient frequency cutoff from the start in order to keep finite the frequency  $\tilde{\omega}_0$  and the shift  $\Delta$ . Contour integration gives  $\tilde{\omega}_0^2 = \omega_0^2 + \sqrt{2}\Gamma_e \Omega^3/(3c)$  and

$$\Delta(\omega) = \frac{2\sqrt{2}}{3\tilde{\omega}_0 c} \Gamma(\omega) \Omega (\Omega^2 + \omega^2). \quad (6.53)$$

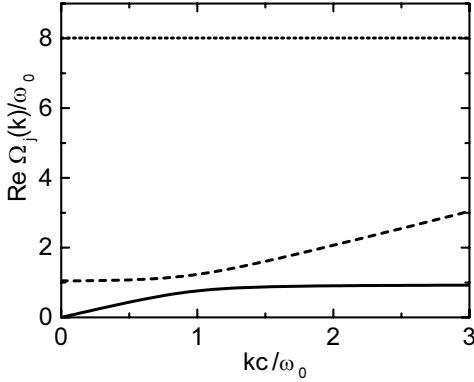
The dielectric function has the form

$$\varepsilon(\omega) = 1 - \frac{4\pi c^2 \Gamma_e n}{\omega^2 - \omega_0^2 + [\sqrt{2}\Omega^3(\Omega^2 - \omega^2)/(3\omega^2 c)] [\Gamma_e - \Gamma(\omega)] + \frac{2}{3}i\Gamma(\omega)\omega^3/c}. \quad (6.54)$$

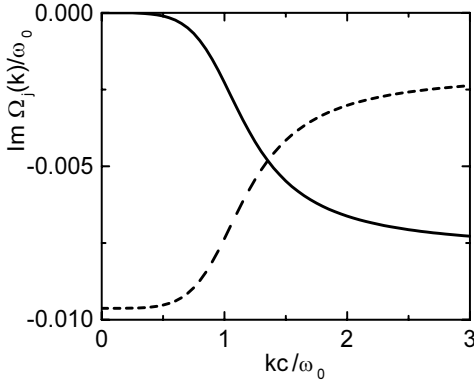
In this case, the resonance frequency shifts to frequencies lower than  $\omega_0$  and the shift is larger for larger cutoff frequencies. However, since the classical electron radius is so much smaller than an optical wavelength, it is very well possible to choose a cutoff frequency such that  $\omega_0 \ll \Omega \ll c/\Gamma_e$ . Then for optical frequencies, the dielectric function (6.54) is of the form (6.51). Note that for high frequencies  $\omega^2 \chi(\omega) \rightarrow -\omega_c^2$  for the dielectric function (6.54), but not for Eq. (6.51).

Again, the frequency cutoff introduces a cutoff branch. In Fig. 6.1 we plot the real parts of the three solutions  $\Omega_j(k)$  of the dispersion relation. As a measure of the damping, we introduce  $\kappa$ , which is given by  $\Gamma_e \omega_0^2/(3c)$ . For the purpose of presentation, the numerical values of both  $\omega_c$  and  $\kappa$  were chosen artificially large for a dilute gas. The frequencies on the cutoff branch are of the same magnitude as the cutoff frequency  $\Omega$ , much higher than the optical regime. The imaginary parts of the upper and lower polariton branches are plotted in Fig. 6.2. The imaginary part of the cutoff branch is large, negative, and practically constant for parameters as given in Fig. 6.1. Again, since the group velocity on the cutoff branch is practically zero, the upper and lower polariton branches together satisfy the sum rules of section 6.3. In particular, Fig. 6.2 illustrates that the upper and lower polariton group velocities  $v_{g,u}$  and  $v_{g,l}$  satisfy the sum rule  $\text{Im}(v_{g,u} + v_{g,l}) = 0$ .

The cutoff, which was necessary to produce the dielectric function in the damped-polariton theory, neatly removes the preacceleration behavior associated with a pole in the upper half plane and leads to a good response function. The form of the coupling  $V(\omega)$  given above Eq. (6.52) has the following physical interpretation. By equating the damped-polariton dielectric function with Eq. (6.51), we assumed that the dilute gas can be described as a homogeneous dielectric. The light scattering by the gas molecules can be accounted for by an absorptive coupling to the *free* electromagnetic field as long as only single scattering of light is relevant. Then scattered light is lost for propagation in the original direction. If the matter-bath coupling is dipole coupling, then for optical frequencies the product  $V^2(\omega_1)/\omega_1$  should be proportional to the density of states of the electromagnetic field, which goes quadratically in frequency. This is indeed the case.

**Figure 6.1:**

Real parts of the three solutions  $\Omega_j$  of the dispersion relation with  $\varepsilon(\omega)$  as in Eq. (6.54). Numerical values of the parameters:  $\Omega = 10\omega_0$ ,  $\omega_c = 0.5\omega_0$  and  $\kappa = 0.01\omega_0$ . The solid line is the lower polariton branch, the upper polariton branch is dashed and the cutoff branch is dotted.

**Figure 6.2:**

Imaginary parts of the lower (solid line) and upper (dashed line) polariton solutions  $\Omega_j$  of the dispersion relation with  $\varepsilon(\omega)$  as in Eq. (6.54). Numerical values of the parameters as in the previous plot. Not shown is the imaginary part of the cutoff branch, which is also negative and about a 1000 times larger in magnitude.

## 6.6 Spontaneous emission

The spontaneous-emission rate, in principle, is a time-dependent quantity. In this section we investigate the transient dynamics of the spontaneous-emission rate of a guest atom in an absorbing medium when the transition frequency of the guest atom is close to a material resonance of the medium. We show how our results relate to previous treatments of spontaneous emission in absorbing dielectrics, where Fermi's golden rule was used to show that the time-independent (equilibrium) value for the spontaneous-emission rate equals  $\Gamma_0 \text{Re}[n(\omega_A)]$  [155, 160]. Recently, local-field effects have been included in quantum electrodynamical formulations of the problem [148, 150, 153, 155, 180], but we shall not focus on them here.

We model the guest atom as a two-level atom with ground state  $|g\rangle$  and excited state  $|e\rangle$  and Hamiltonian  $H_A = \hbar\omega_A|e\rangle\langle e|$ . The medium (with fields and bath included) is described by the damped-polariton model with Hamiltonian  $H_M$  given by Eq. (6.1). The total Hamiltonian is  $H = H_0 + V$  with  $H_0 = H_M + H_A$  and  $V = -\boldsymbol{\mu}_A \cdot \mathbf{E}(\mathbf{r}_A)$ . Here  $V$  is the dipole interaction between the atom and the medium,  $\boldsymbol{\mu}_A$  is the atomic dipole moment operator, and  $\mathbf{E}(\mathbf{r}_A)$  is the electric field operator at the position  $\mathbf{r}_A$  of the atom.

Suppose that the damped-polariton system is prepared at time 0 in a state described

by a density matrix  $\rho_M(0)$ . We do not assume that  $\rho_M(0)$  commutes with  $H_M$  nor that it factorizes into a product of a density operator for the bath and a density operator for the undamped-polariton system (as is often assumed for convenience [181]). At time  $t_0 > 0$  we bring the guest atom in its excited state and couple it to the damped-polariton system. Using perturbation theory, one can calculate [45] the time-dependent probability that the guest atom has emitted a photon at time  $t > t_0$ . We define the derivative of this quantity as the instantaneous spontaneous-emission rate  $\Gamma(t)$ . It is given as

$$\Gamma(t) = \frac{2}{\hbar^2} \text{Re} \int_{t_0}^t dt' e^{i\omega_A(t-t')} \text{Tr}[\rho_M(0) \boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{r}_A, t) \boldsymbol{\mu} \cdot \mathbf{E}(\mathbf{r}_A, t')], \quad (6.55)$$

where  $\boldsymbol{\mu}$  is now the dipole transition matrix element of the guest atom.

If the guest atom is excited a long time after the initial preparation of the medium, all transient effects in the electric field have damped out. Hence, the field may be replaced by its long-time limit  $\mathbf{E}_l(\mathbf{r}_A, t)$ , which is given in Eq. (6.34). Since  $\mathbf{E}_l$  depends only on the bath operators at  $t = 0$ , we may write Eq. (6.55) in the form

$$\Gamma(t) = \frac{2}{\hbar^2} \text{Re} \int_{t_0}^t dt' e^{i\omega_A(t-t')} \text{Tr}_{\text{bath}}[\rho_{\text{red}}(0) \boldsymbol{\mu} \cdot \mathbf{E}_l(\mathbf{r}_A, t) \boldsymbol{\mu} \cdot \mathbf{E}_l(\mathbf{r}_A, t')]. \quad (6.56)$$

Here  $\rho_{\text{red}}$  is the reduced density matrix obtained by tracing out the electromagnetic and material degrees of freedom:  $\rho_{\text{red}}(0) = \text{Tr}_{\text{em,mat}} \rho_M(0)$ . For the special case that the initial density matrix  $\rho_M(0)$  factorizes, the reduced density matrix is the bath density matrix  $\rho_{\text{bath}}(0)$  at  $t = 0$ . In general, the initial state of the electromagnetic and material degrees of freedom at  $t = 0$  does not play a role in the emission rate.

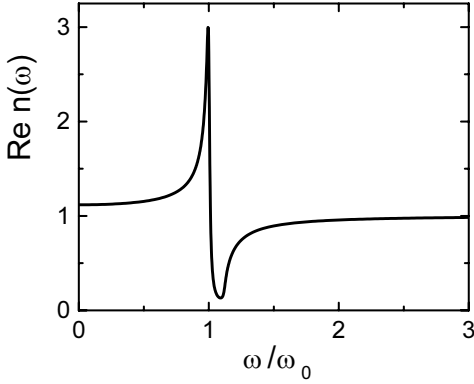
Spontaneous emission in its pure form arises if the reduced density matrix describes the ground state of the bath. Let us assume this is indeed the case. Upon inserting Eq. (6.34) in Eq. (6.56) we can perform the  $t'$  integral, the integrals over the wavevector, and the summations over the polarization directions. This leads to

$$\Gamma(t) = \frac{\mu^2}{3\pi^2 \hbar \epsilon_0 c^3} \text{Re} \int_0^\infty d\omega \omega^3 n(\omega) \frac{\sin[(\omega - \omega_A)(t - t_0)]}{\omega - \omega_A}, \quad (6.57)$$

with  $n(\omega) = \sqrt{\epsilon(\omega)}$  the complex refractive index.

For times  $(t - t_0)$  that are large enough, one may replace  $\sin[(\omega - \omega_A)(t - t_0)] / (\omega - \omega_A)$  by  $\pi \delta(\omega - \omega_A)$ . However, the time scale at which this replacement is valid depends on the resonance structure of the refractive index  $n(\omega)$ . Since we want to study just this time scale, we will not make the replacement. To evaluate the integral, we multiply the integrand by a convergence factor  $\Omega^4 / (\Omega^4 + \omega^4)$  with  $\Omega \gg \omega_A$ . The specific choice of the cutoff frequency  $\Omega$  will only affect  $\Gamma(t)$  at time differences  $t - t_0$  much smaller than a single optical cycle. We need to use a high-frequency cutoff at this point because the dipole approximation is incorrect for high frequencies.

For the dielectric function, we take the Lorentz oscillator form (6.50) and we choose the cutoff frequency in that model to be identical to the one inserted in Eq. (6.57). In Fig. 6.3, we give the real part of the refractive index that clearly changes rapidly near  $\omega = \omega_0$ . It is a familiar figure and it shows that the refractive index does not change much

**Figure 6.3:**

Real part of the refractive index in the Lorentz oscillator model when  $\varepsilon(\omega)$  is given by Eq. (6.50) with parameters  $\kappa_0 = 0.01\omega_0$ ,  $\omega_c = 0.5\omega_0$ , and  $\Omega = 10\omega_0$ .

while increasing the cutoff frequency  $\Omega$  from  $10\omega_0$  to infinity. The density of radiative modes around the material resonance is proportional to  $\omega^2 \text{Re}[n(\omega)]$ .

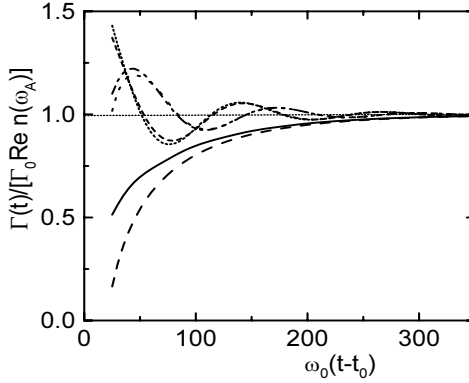
With this model for the dielectric function and the parameters as in Fig. 6.3, we calculated  $\Gamma(t)$  in the case that the transition frequency  $\omega_A$  exactly equals  $\omega_0$ . Since the integrand in Eq. (6.57) is rapidly fluctuating, it is expedient to use complex contour deformation to evaluate the integral. We add an infinitesimal positive imaginary part to the denominator and split the sine into two complex exponentials. The contour of the integral with  $\exp[i(\omega - \omega_A)(t - t_0)]$  in the integrand is deformed towards the positive imaginary axis. The contribution from the pole arising from the convergence factor can be neglected at time scales  $t - t_0 \gg \omega_A^{-1}$ . Likewise, the integration contour of the integral with  $\exp[-i(\omega - \omega_A)(t - t_0)]$  is deformed towards the negative imaginary axis. Again, the pole contribution from the convergence factor is negligible. Further contributions, which cannot be neglected, arise from the branch cuts of  $n(\omega) = \sqrt{\varepsilon(\omega)}$  and from the pole at  $\omega_A$ . The latter contribution yields the equilibrium value  $\Gamma(\infty) = \Gamma_0 \text{Re } n(\omega_A)$ . In contrast, the branch cuts yield time-dependent contributions to  $\Gamma(t)$ . For large  $\Omega$  they are situated at  $\omega_1 = -i\kappa_0 + \sqrt{\omega_0^2 - \kappa_0^2}$  and  $\omega_2 = -i\kappa_0 + \sqrt{\omega_0^2 + \omega_c^2 - \kappa_0^2}$ . Around  $\omega_1$  and  $\omega_2$ , we can approximate the dielectric function by

$$\varepsilon(\omega_1 - i\delta e^{i\varphi}) \simeq -i\omega_c^2 e^{-i\varphi} / \left( 2\delta \sqrt{\omega_0^2 - \kappa_0^2} \right), \quad (6.58a)$$

$$\varepsilon(\omega_2 - i\delta e^{i\varphi}) \simeq -2i\delta e^{i\varphi} \sqrt{\omega_0^2 + \omega_c^2 - \kappa_0^2} / \omega_c^2. \quad (6.58b)$$

The branch cut at  $\omega_1$  gives the following contribution to the spontaneous-emission rate:

$$-\frac{\Gamma_0}{\pi\omega_A^3} \text{Re} \left[ \exp\{-i\pi/4 - \kappa_0(t - t_0) - i(\sqrt{\omega_0^2 - \kappa_0^2} - \omega_A)(t - t_0)\} J(t) \right], \quad (6.59)$$

**Figure 6.4:**

Normalized spontaneous-emission rate  $\Gamma(t)/[\Gamma_0 \text{Re } n(\omega_A)]$  in an absorbing dielectric as a function of time. Choice of parameters in the Lorentz oscillator model:  $\kappa_0 = 0.01\omega_0$ ,  $\omega_c = 0.5\omega_0$ ,  $\Omega = \infty$ . Results are presented for three transition frequencies  $\omega_A$ : for  $\omega_A/\omega_0 = 1.0$ , the solid line represents the exact numerical result for Eq. (6.57) and the dashed line is the approximate analytical expression Eq. (6.61). For  $\omega_A/\omega_0 = 0.95$ , the dotted line is exact and the dash-dot-dotted line is the approximation. The exact result for  $\omega_A/\omega_0 = 1.05$  is the short-dashed line and the short-dotted line is its approximation.

where  $J(t)$  is defined as:

$$J(t) = \int_0^\infty d\lambda \frac{e^{-\lambda(t-t_0)} \left[ \sqrt{\omega_0^2 - \kappa_0^2} - i(\lambda + \kappa_0) \right]^3}{\sqrt{\lambda} \left[ \sqrt{\omega_0^2 - \kappa_0^2} - \omega_A - i(\lambda + \kappa_0) \right]} \times \left[ \frac{2\omega_c^2 \sqrt{\omega_0^2 - \kappa_0^2} + i\lambda(\lambda^2 + 4\omega_0^2 + \omega_c^2 - 4\kappa_0^2)}{4(\omega_0^2 - \kappa_0^2) + \lambda^2} \right]^{1/2}. \quad (6.60)$$

The branch cut around  $\omega_2$  gives a similar contribution.

The integrals arising from the branch cuts and from the imaginary axis can be evaluated numerically since their integrands are no longer rapidly fluctuating. The on-resonance result  $\omega_A = \omega_0$  is the solid line in Fig. 6.4. We see that the spontaneous-emission rate builds up until it finally reaches the time-independent equilibrium value  $\Gamma_0 \text{Re } n(\omega_A)$ .

The dashed line in Fig. 6.4 is an analytical approximation for  $\Gamma(t)$ , which captures the main features of the on-resonance time dependence at least qualitatively. It is derived by retaining only the contribution (6.59) in the time-dependent part of  $\Gamma(t)$  as this is dominant for large  $t$ . Moreover, we approximate  $J(t)$  by the first term in its asymptotic expansion for large  $t - t_0$ . In this way we arrive at the following approximate expression for  $\Gamma(t)$ :

$$\Gamma(t) \simeq \Gamma_0 \text{Re } [n(\omega_A)] - \Gamma_0 \text{Re} \left[ \frac{\omega_c (\omega_0^2 - \kappa_0^2)^{5/4} \exp\{-i\pi/4 - (i\Delta - \kappa_0)(t - t_0)\}}{\sqrt{2\pi} \omega_A^3 (\Delta - i\kappa_0)(t - t_0)^{1/2}} \right], \quad (6.61)$$

where  $\Delta$  was used as abbreviation for  $\sqrt{\omega_0^2 - \kappa_0^2} - \omega_A$ . As explained, this approximation contains only the contribution from the branch cut at  $\omega_1$ ; the branch cut at  $\omega_2$  gives a

faster decaying term, which goes like  $e^{-\kappa_0(t-t_0)}/(t-t_0)^{3/2}$ . The contributions from the integrals along the imaginary axis decay even faster.

It can be seen from Eq. (6.61) that the amplitude of the time-dependent part of  $\Gamma(t)$  falls off as  $e^{-\kappa_0(t-t_0)}/(t-t_0)^{1/2}$  and also that the amplitude of the extra term is largest around resonance when  $\omega_A \simeq \sqrt{\omega_0^2 - \kappa_0^2}$ . Away from resonance, oscillations with frequency  $\sqrt{\omega_0^2 - \kappa_0^2} - \omega_A$  are present. Fig. 6.4 also shows the two off-resonance cases  $\omega_A = 0.95\omega_0$  and  $\omega_A = 1.05\omega_0$ , where both the exact numerical decay rates and their analytical approximations show time-dependent oscillations.

The main result of the present discussion is the time dependence of the spontaneous-emission rate. The time-independent value is not reached instantaneously, but at a time scale that is governed by the resonance characteristics of the medium. In fact, the smaller the resonance width  $\kappa_0$ , the longer it takes to reach the time-independent value. Typically, it takes  $\omega_0/\kappa_0$  optical cycles as follows from the exponential  $e^{-\kappa_0(t-t_0)}$  in the approximate expression (6.61). For narrow resonances with  $\omega_0/\kappa_0$  large, the transient dynamics may take a substantial amount of time.

## 6.7 Discussion and conclusions

We have solved the equations of motion for the field operators in the damped-polariton model using Laplace transformations. The solutions of the field and medium operators are the sum of a transient and a permanent part. The latter are expressed solely in terms of the initial bath operators. Long after the initial time, all field and medium operators are functions of the bath operators alone provided the coupling to the bath is nonzero for all frequencies. The long-time solutions satisfy quantum Langevin equations in which the initial bath operators figure as the quantum noise source. The same continuum that produces the absorption also forms the noise source that keeps the commutation relations in order. This is conceptually simpler than expressing the quantum Langevin noise in terms of the creation and annihilation operators that diagonalize the total Hamiltonian of the damped-polariton model [158].

The effects of the initial state of the field and medium variables on the expectation values at a later time are noticeable only during a short period that is determined by the characteristic relaxation times of the damped-polariton modes. Once these transient effects have died out the expectation values are determined by the reduced density matrix, which follows from the full density matrix at the initial time by taking the trace over the degrees of freedom of field and matter (without bath). If the full density matrix at the initial time factorizes, the reduced density matrix equals the initial bath density matrix.

The method of long-time solutions can be used for other dissipative quantum systems as well. For models in which the Hamiltonian can be diagonalized completely, it is an alternative to the Fano diagonalization technique [182]. The latter can be quite complicated [158, 183], whereas our long-time solutions are found after the simple inversion of a  $4 \times 4$  matrix, as one sees from section 6.2 and 6.4. More generally, the long-time method may be useful for dissipative systems with a bilinear coupling to a harmonic oscillator bath whose dynamics can be integrated out.

We employed the method of long-time solutions to study transient effects in a medium

described by a Lorentz oscillator dielectric function. This dielectric function (and that of the point-scattering model as well) can be derived from the damped-polariton model by taking a suitable bath coupling. Although a cutoff procedure turns out to be indispensable, the essential physics in the optical regime can be represented adequately in this way. Once the connection with the damped-polariton model has been established, spontaneous-emission processes by a guest atom in a Lorentz oscillator dielectric can be investigated by means of the long-time method. Although transient effects due to the initial preparation of the dielectric have damped out after a few medium relaxation periods, transient behavior of a different type shows up in the initial stages of the decay process. This transient behavior, which is related to the preparation of the guest atom in its excited state, leads to a nonexponential decay - or in other words to a time-dependent spontaneous-emission rate - if the atomic transition frequency is near a resonance of the dielectric. The nonexponential dynamics takes place at time scales that are inversely proportional to the width of the resonance. As we have shown, the characteristics of the time-dependent decay rate can be captured in an analytic asymptotic expression of which the qualitative features are corroborated by numerical methods.



## Appendix A

# Analytical expression for T-matrix of N-plane crystal

*Inversion.* The problem that arose in section 2.3.2 and which is solved in this appendix is how to find the inverse of the matrix

$$\mathbf{M} \equiv \begin{pmatrix} \nu & x & x^2 & x^3 \\ x & \nu & x & x^2 \\ x^2 & x & \nu & x \\ x^3 & x^2 & x & \nu \end{pmatrix}. \quad (\text{A.1})$$

The solution will be for general  $N$ , but matrices are presented for  $N = 4$ . Define the upper diagonal matrix  $\mathbf{U}$  and lower diagonal matrix  $\mathbf{L}$  as

$$\mathbf{U} = \begin{pmatrix} 0 & x & 0 & 0 \\ 0 & 0 & x & 0 \\ 0 & 0 & 0 & x \\ 0 & 0 & 0 & 0 \end{pmatrix}; \quad \mathbf{L} = \begin{pmatrix} 0 & 0 & 0 & 0 \\ x & 0 & 0 & 0 \\ 0 & x & 0 & 0 \\ 0 & 0 & x & 0 \end{pmatrix}. \quad (\text{A.2})$$

Then we have

$$\mathbf{U}^2 = \begin{pmatrix} 0 & 0 & x^2 & 0 \\ 0 & 0 & 0 & x^2 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}; \quad \mathbf{U}^N = 0, \quad (\text{A.3})$$

and similarly for powers of the lower diagonal matrix  $\mathbf{L}$ . From this, it follows that we can write the matrix  $\mathbf{M}$  as

$$\begin{aligned} \mathbf{M} &= \nu \mathbf{1} + \sum_{p=1}^{N-1} (\mathbf{U}^p + \mathbf{L}^p) \\ &= \nu \mathbf{1} + \mathbf{U}(\mathbf{1} - \mathbf{U})^{-1} + (\mathbf{1} - \mathbf{L})^{-1} \mathbf{L}. \end{aligned} \quad (\text{A.4})$$

Now consider the inverse of the matrix product  $(\mathbb{1} - \mathbf{L})\mathbf{M}(\mathbb{1} - \mathbf{U})$ . Use this and equation (A.4) to arrive at the following expression for the inverse of  $\mathbf{M}$ :

$$\mathbf{M}^{-1} = (\mathbb{1} - \mathbf{U}) [\nu(\mathbb{1} - \mathbf{L})(\mathbb{1} - \mathbf{U}) + (\mathbb{1} - \mathbf{L})\mathbf{U} + \mathbf{L}(\mathbb{1} - \mathbf{U})]^{-1} (\mathbb{1} - \mathbf{L}). \quad (\text{A.5})$$

In this form, finding the inverse is much easier than before, because the matrix  $[\dots]$  has only nonzero elements on the diagonal and first off-diagonals. This becomes clearer by rewriting

$$\mathbf{M}^{-1} = (\mathbb{1} - \mathbf{U}) [\mathbf{Y} + \mathbf{Z}]^{-1} (\mathbb{1} - \mathbf{L}) = (\mathbb{1} - \mathbf{U}) [\mathbb{1} + \mathbf{Y}^{-1}\mathbf{Z}]^{-1} \mathbf{Y}^{-1} (\mathbb{1} - \mathbf{L}), \quad (\text{A.6})$$

where  $\mathbf{Y} \equiv (\nu + \nu x^2 - 2x^2)\mathbb{1} + (1 - \nu)(\mathbf{U} + \mathbf{L})$  is a symmetric band diagonal matrix and the matrix  $\mathbf{Z}$  has matrix elements  $Z_{kl} \equiv (2 - \nu)x^2\delta_{k1}\delta_{l1}$  so that only its upper left element is nonzero.

The matrix  $\mathbf{M}^{-1}$  is known when  $\mathbf{Y}^{-1}$  and  $[\mathbb{1} + \mathbf{Y}^{-1}\mathbf{Z}]^{-1}$  are known. Now  $\mathbf{Y}$  has a simple structure and it can be inverted immediately using the transformation matrix  $J_{kl} \equiv \sqrt{2/(N+1)} \sin[kl\pi/(N+1)]$ . It is its own inverse and it diagonalizes  $\mathbf{Y}$ : we have  $\mathbf{Y}^{-1} = \mathbf{J}\mathbf{\Lambda}^{-1}\mathbf{J}$  with  $\Lambda_{kl}^{-1} = \delta_{kl}f^{-1}(k)$  where the function  $f(k)$  is defined as

$$f(k) \equiv \nu(1 + x^2) - 2x^2 + 2x(1 - \nu) \cos[k\pi/(N+1)] \quad (k = 1, 2, \dots, N). \quad (\text{A.7})$$

What remains to be done for the inversion of  $\mathbf{M}$  is the evaluation of the infinite series

$$(\mathbb{1} + \mathbf{Y}^{-1}\mathbf{Z})^{-1} = \mathbb{1} - \mathbf{Y}^{-1}\mathbf{Z} + (\mathbf{Y}^{-1}\mathbf{Z})^2 - (\mathbf{Y}^{-1}\mathbf{Z})^3 + \dots \quad (\text{A.8})$$

Now since we have

$$(Y^{-1}Z)_{kp} = (2 - \nu)x^2\delta_{1p} \sum_{m=1}^N J_{km}f^{-1}(m)J_{mp} \equiv h_N(k, p)\delta_{1p}, \quad (\text{A.9})$$

where in the last equality the function  $h_N(k, p)$  was defined, we find that  $[(\mathbf{Y}^{-1}\mathbf{Z})^n]_{kp} = h_N(k, 1)h_N^{n-1}(1, 1)\delta_{1p}$ . Summing up all orders, we find that

$$(\mathbb{1} + Y^{-1}Z)_{kp}^{-1} = \left[ \delta_{kp} - \left( \frac{h_N(k, 1)}{1 + h_N(1, 1)} \right) \delta_{1p} \right]. \quad (\text{A.10})$$

Inserting this into equation (A.6), we have found the inverse of the matrix  $\mathbf{M}$ . This result is used in equation (2.30) of section 2.3.2.

*Symmetry.* The T-matrix elements should have the symmetry  $T_{\alpha, \beta}^{(N)} = T_{N+1-\alpha, N+1-\beta}^{(N)}$ , but in the expression (2.30) for the T-matrix that was used in the calculations, this symmetry is not manifest, as was remarked in section 2.5. Here we show how the hidden symmetry can be pulled out.

All terms in Eq. (2.30) should be brought under one denominator. Rewrite the sines and cosines in the expression in terms of complex exponentials and introduce new variables for the exponentials such that the expression becomes a polynomial in the new variables. In this form it is easier to find that the nominator and denominator have the common

factors  $[x - \exp(iKa)]$  and  $[x - \exp(-iKa)]$  that can be divided out. For the T-matrix we then find

$$T_{\alpha\beta}^{(N)} = \frac{ik_z [x^2 - 2x \cos(Ka) + 1]^2}{x(x^2 - 1) \sin(Ka)} H_{\alpha\beta}^{(N)}, \quad (\text{A.11})$$

with  $H_{\alpha\beta}^{(N)}$  for  $\alpha > \beta$  defined as

$$\begin{aligned} H_{\alpha\beta}^{(N)} &\equiv \frac{\{x \sin[(\alpha - N)Ka] - \sin[(\alpha - N - 1)Ka]\}}{\{x \cos[(N - 1)Ka/2] - \cos[(N + 1)Ka/2]\}} \times \\ &\times \frac{\{x \sin[(\beta - 1)Ka] - \sin[\beta Ka]\}}{\{x \sin[(N - 1)Ka/2] - \sin[(N + 1)Ka/2]\}}. \end{aligned} \quad (\text{A.12})$$

For  $\alpha < \beta$  the  $\alpha$  and  $\beta$  should be interchanged in this expression. When  $\alpha = \beta$ , it can be shown that an  $\alpha$ -independent term  $\Delta H_{\alpha\alpha}$  should be added to the expression for  $H_{\alpha\beta}$ :

$$\begin{aligned} \Delta H_{\alpha\alpha} &= \frac{-x[x^2 - 2x \cos(Ka) + 1]^{-1} [x \cos(Ka) - 1]^2}{\{x \cos[(N - 1)Ka/2] - \cos[(N + 1)Ka/2]\}} \times \\ &\times \frac{\{x^2 \sin[(N - 1)Ka] - 2x \sin(NKa) + \sin[(N + 1)Ka]\}}{\{x \sin[(N - 1)Ka/2] - \sin[(N + 1)Ka/2]\}}. \end{aligned} \quad (\text{A.13})$$

From Eqs. (A.12) and (A.13) we can infer that  $H_{\alpha\beta}^{(N)} = H_{N+1-\alpha, N+1-\beta}^{(N)}$ , where it is important to notice that  $N + 1 - \alpha < N + 1 - \beta$  when  $\alpha > \beta$ . It follows that also the T-matrix has the required symmetry:  $T_{\alpha\beta}^{(N)} = T_{N+1-\alpha, N+1-\beta}^{(N)}$ .

*Determinant.* For section 2.4.2 it is useful to compute the determinant of the matrix  $\mathbf{M}$  and this can be done best using equation (A.6):  $\det(\mathbf{M}) = \det[\mathbf{Y}(\mathbf{1} + \mathbf{Y}^{-1}\mathbf{Z})]$ . This determinant is equal to  $[1 + h_N(1, 1)]$  times the product of all the  $f(k)$ 's of equation (A.7). With equation (2.32) and the definition of the Bloch wave vector this gives

$$\begin{aligned} \det[\mathbf{M}(k_{\parallel}, \omega)] &= \left[ \frac{-1}{2x(1 - \nu)} \right]^N \left\{ \sin[(N + 1)Ka] - \left[ \frac{(2 - \nu)x}{1 - \nu} \right] \sin[NKa] \right\} \\ &\times \frac{\prod_{m=1}^N \left[ \cos(Ka) - \cos\left(\frac{m\pi}{N+1}\right) \right]}{\sin[(N + 1)Ka]}. \end{aligned} \quad (\text{A.14})$$

Notice that the zeroes of the determinant are *not* caused by the products of  $\cos(Ka) - \cos\left(\frac{m\pi}{N+1}\right)$  going to zero, because of the  $\sin[(N + 1)Ka]$  in the denominator. The determinant is zero when the remaining factor in the nominator is zero, which is equivalent to  $[1 + h_N(1, 1)] = 0$ .



## Appendix B

# Functional differentiation after choosing a gauge

After choosing the gauge, the vector potential is generalized transverse. The Euler-Lagrange equation for the vector potential is therefore an equation of motion of a constrained system, where the constraint is the gauge condition (4.4). One could try and solve this problem using the method of Lagrange multipliers [184], but this is not the route that will be pursued here. Instead, the appropriate definition and computation of functional derivatives after choosing a gauge will be studied in sections B.1 and B.2, respectively.

### B.1 Two definitions of functional derivatives

Before choosing the gauge, the functional derivative of the Lagrangian with respect to the vector potential is defined as

$$\frac{\delta L}{\delta \mathbf{A}(\mathbf{r})} \equiv \lim_{\gamma \rightarrow 0} \frac{\int d\mathbf{r}' \{ \mathcal{L}[\mathbf{A}(\mathbf{r}') + \gamma \delta(\mathbf{r} - \mathbf{r}') \mathbf{I}] - \mathcal{L}[\mathbf{A}(\mathbf{r}')]\}}{\gamma}. \quad (\text{B.1})$$

The functional derivative of the functional  $L$  with respect to the function  $\mathbf{A}$  describes the relative changes of  $L$  when small variations proportional to  $\delta(\mathbf{r} - \mathbf{r}')\mathbf{I}$  are added to the function  $\mathbf{A}$ . It turns out that the right-hand side of Eq. (B.1) can be computed as the partial derivative of the Lagrangian density with respect to  $\mathbf{A}$ . While doing this, the Lagrangian density  $\mathcal{L}$  can simply be considered as a function and  $\mathbf{A}$  as one of its variables. Before choosing the gauge, the functional derivative so defined leads to the third and fourth Maxwell equations.

After choosing the gauge, the interaction term  $L_{\mathbf{A}\Phi}$  (4.37) becomes identically zero, as noted in the main text. However, its functional derivative as defined in (B.1) with respect to  $\dot{\mathbf{A}}(\mathbf{r})$  leads to the nonzero answer  $\varepsilon_0 \varepsilon(\mathbf{r}) \nabla \Phi(\mathbf{r})$ . An identically zero function should not have a nonzero derivative. This can only mean that Eq. (B.1) does not define the functional derivative with respect to generalized transverse functions correctly. The reason is that

the function space in which the vector potential lives has become smaller by choosing the gauge: it now lives in the subspace of functions which are generalized transverse. This also means that functional variations of  $\mathbf{A}$  should stay inside this subspace. In the functional derivative (B.1), variations in the whole function space are allowed and clearly  $\nabla_{\mathbf{r}'} \cdot [\varepsilon(\mathbf{r}')\delta(\mathbf{r} - \mathbf{r}')\mathbf{I}]$  is nonzero.

With every set of constraints a new functional derivative can be associated. Here, only the gauge-constraint will be considered that functions be generalized transverse. Functional differentiation with respect to generalized transverse functions can be defined as [86]

$$\frac{\delta L}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \equiv \lim_{\gamma \rightarrow 0} \frac{\int d\mathbf{r}' \{ \mathcal{L}[\mathbf{A}(\mathbf{r}') + \gamma \delta_\varepsilon^T(\mathbf{r}', \mathbf{r})] - \mathcal{L}[\mathbf{A}(\mathbf{r}')]\}}{\gamma}, \quad (\text{B.2})$$

with the generalized transverse delta function  $\delta_\varepsilon^T$  as defined in Eq. (4.12). In this new functional derivative, the functional variations do stay inside the generalized transverse subspace, since  $\nabla_{\mathbf{r}'} \cdot [\varepsilon(\mathbf{r}')\delta_\varepsilon^T(\mathbf{r}', \mathbf{r})] = 0$ . It will be called the “constrained functional derivative” in the following. Note that the constrained functional derivative must also be used in free space after choosing the Coulomb gauge (see [84], p. 289); for free space the generalized transverse delta function reduces to the free-space transverse delta function  $\delta^T$ , given in Eq. (C.5).

## B.2 Simple rules to compute constrained functional derivatives

Now the goal is to find simple rules to compute the constrained functional derivative (B.2) with respect to generalized transverse functions, just like the normal functional derivative (B.1) can simply be calculated as a partial derivative. With that goal in mind, note with the use of Eqs. (4.15a) and (4.15d) that the functional derivative (B.2) is simple in the following two important cases:

$$\frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \mathbf{X}^T(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') = \mathbf{X}^T(\mathbf{r}), \quad (\text{B.3})$$

$$\frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \varepsilon(\mathbf{r}') \mathbf{X}^L(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') = 0, \quad (\text{B.4})$$

where  $\mathbf{X}^T$  and  $\mathbf{X}^L$  are arbitrary transverse and longitudinal functions, respectively. The second important case makes clear that the constrained functional derivative of zero (inner products such as  $L_{\mathbf{A}\Phi}$  of transverse and longitudinal fields) indeed gives zero; the first important case (B.3) shows that the partial-derivative-of- $\mathcal{L}$  computation rule still gives the correct answers for inner products of  $\mathbf{A}_\varepsilon^T$  with transverse functions. In combination with the definition (B.2) of the constrained functional derivative, this justifies the naive calculation of the canonical field in Eq. (4.7) of section 4.2:

$$\frac{\delta}{\delta \dot{\mathbf{A}}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \varepsilon(\mathbf{r}') \dot{\mathbf{A}}^2(\mathbf{r}') = 2 \varepsilon(\mathbf{r}) \dot{\mathbf{A}}(\mathbf{r}). \quad (\text{B.5})$$

How can the constrained functional derivative be calculated in the more general situation

$$\frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \mathbf{X}(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}'), \quad (\text{B.6})$$

where  $\mathbf{X}$  is a general vector function? It will now be shown that any vector field  $\mathbf{X}$  can be decomposed such that the only two rules of computation needed are the simple cases (B.3) and (B.4).

Given the vector field  $\mathbf{X}$ , construct the scalar field  $\sigma_{\mathbf{X}} = -\nabla \cdot \mathbf{X}$ . Now find the potential  $\chi$ , given the “charge distribution”  $\sigma_{\mathbf{X}}$  and the dielectric function  $\varepsilon_0 \varepsilon(\mathbf{r})$ , from the following generalized Poisson equation:

$$\varepsilon_0 \nabla \cdot [\varepsilon(\mathbf{r}) \nabla \chi(\mathbf{r})] = -\sigma_{\mathbf{X}}(\mathbf{r}). \quad (\text{B.7})$$

This is a well-known problem in electrostatics. There is a unique solution for  $\chi$  of this inhomogeneous problem, given the charge distribution and the boundary condition that the potential be zero at infinity. With the potential  $\chi$  thus found, define two vector fields  $\mathbf{X}_1$  and  $\mathbf{X}_2$  as

$$\mathbf{X}_1 \equiv \mathbf{X}(\mathbf{r}) - \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \chi(\mathbf{r}), \quad (\text{B.8})$$

$$\mathbf{X}_2 \equiv \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \chi(\mathbf{r}), \quad (\text{B.9})$$

so that evidently  $\mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2$ . The vector field  $\mathbf{X}_1$  is transverse by construction of the potential  $\chi$ ; the field  $[\mathbf{X}_2/\varepsilon(\mathbf{r})]$  is of course longitudinal. In summary, the following theorem was proven: *An arbitrary vector field can be uniquely decomposed into a part which after division by  $\varepsilon(\mathbf{r})$  is longitudinal, and a transverse part.* This theorem is useful for evaluating the constrained functional derivative, because

$$\begin{aligned} \frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \mathbf{X}(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') &= \frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' [\mathbf{X}_1(\mathbf{r}') + \mathbf{X}_2(\mathbf{r}')] \cdot \mathbf{A}(\mathbf{r}') \\ &= \mathbf{X}(\mathbf{r}) - \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \chi(\mathbf{r}), \end{aligned} \quad (\text{B.10})$$

where the simple derivatives Eqs. (B.3) and (B.4) were used. The problem of computing a functional derivative with respect to a generalized transverse function has thus been reduced to a problem in electrostatics. Note that the constrained functional derivative (B.10) produces a field that is always transverse. That transverse field is equal to the transverse part of  $\mathbf{X}$  if  $\mathbf{X}$  itself is transverse or if  $\varepsilon(\mathbf{r}) \equiv 1$ .

As a corollary of the unique decomposition (B.8) and (B.9) of vector fields, a new and short proof can be given of the “generalized Helmholtz theorem” [14], which states that *Every vector field  $\mathbf{Y}$  can be uniquely decomposed as the sum of a generalized transverse vector field  $\mathbf{Y}_1$  and a longitudinal field  $\mathbf{Y}_2$ .* The proof is simple: given  $\mathbf{Y}$ , define  $\mathbf{X} = \varepsilon(\mathbf{r})\mathbf{Y}$ . Then apply the previous unique decomposition to  $\mathbf{X}$ , as in Eqs. (B.8) and (B.9). Define  $\mathbf{Y}_1 \equiv \mathbf{X}_1/\varepsilon(\mathbf{r})$  and  $\mathbf{Y}_2 \equiv \mathbf{X}_2/\varepsilon(\mathbf{r})$ . Then it follows that  $\mathbf{Y} = \mathbf{Y}_1 + \mathbf{Y}_2$ , where  $\mathbf{Y}_1$  is a generalized transverse and  $\mathbf{Y}_2$  is a longitudinal field. This completes the proof.

### B.3 Functional derivatives of the minimal-coupling Lagrangian

In the special case that the vector field  $\mathbf{X}$  is the current density  $\mathbf{J}_g$  produced by the guest atoms in the dielectric, equation (4.34) shows that the “charge density”  $\sigma_{\mathbf{J}_g}$  equals the time-derivative of the physical charge density  $\sigma_g$ , and by the uniqueness of the solution of the generalized Poisson equation, the potential  $\chi$  must be identified with the time-derivative of the physical scalar potential  $\Phi$ . Therefore, the constrained functional derivative of  $\int d\mathbf{r} \mathbf{J}_g \cdot \mathbf{A}$  can now be computed as

$$\frac{\delta}{\delta \mathbf{A}_\varepsilon^T(\mathbf{r})} \int d\mathbf{r}' \mathbf{J}_g(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') = \mathbf{J}_g(\mathbf{r}) - \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \dot{\Phi}(\mathbf{r}). \quad (\text{B.11})$$

With this result, the Euler-Lagrange equation for the vector potential from the Lagrangian (4.39) becomes:

$$\mu_0^{-1} \nabla \times \nabla \times \mathbf{A} + \varepsilon_0 \varepsilon(\mathbf{r}) \ddot{\mathbf{A}} = \mathbf{J}_g - \varepsilon_0 \varepsilon(\mathbf{r}) \nabla \dot{\Phi}. \quad (\text{B.12})$$

This is precisely the equation that was needed for the vector potential such that the fourth Maxwell equation holds also in the generalized Coulomb gauge.

## Appendix C

# Dyadic Green and delta functions

The (full) Green tensor  $\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega)$  of an inhomogeneous medium characterized by the dielectric function  $\varepsilon(\mathbf{r})$  is the solution of the wave equation

$$-\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) + \varepsilon(\mathbf{r})(\omega/c)^2 \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}')\mathbf{I}, \quad (\text{C.1})$$

where the right-hand side is the ordinary Dirac delta function times the unit tensor. Another useful Green function (which actually is a tensor as well) can be found by projecting out the left and right-hand sides of this equation with the generalized transverse delta function (4.12). In doing so, the transverse double-curl term is projected onto itself, see Eq. (4.15a). The (full) Green function can therefore be uniquely projected onto its generalized transverse part  $\mathbf{G}^T$  that is the solution of

$$-\nabla \times \nabla \times \mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) + \varepsilon(\mathbf{r})(\omega/c)^2 \mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) = \bar{\delta}_\varepsilon^T(\mathbf{r}', \mathbf{r}). \quad (\text{C.2})$$

The bar in  $\bar{\delta}_\varepsilon^T$  denotes the transpose. The longitudinal Green function  $\mathbf{G}^L$  is now defined as the difference between  $\mathbf{G}$  and  $\mathbf{G}^T$ , and has the form

$$\mathbf{G}^L(\mathbf{r}, \mathbf{r}') \equiv \frac{1}{(\omega/c)^2 \varepsilon(\mathbf{r})} [\delta(\mathbf{r} - \mathbf{r}')\mathbf{I} - \bar{\delta}_\varepsilon^T(\mathbf{r}', \mathbf{r})] \equiv \frac{1}{(\omega/c)^2 \varepsilon(\mathbf{r})} \bar{\delta}_\varepsilon^L(\mathbf{r}', \mathbf{r}). \quad (\text{C.3})$$

In the last equality of (C.3) the generalized longitudinal delta function was defined as the difference between the ordinary Dirac and the generalized transverse delta, so that  $\bar{\delta}_\varepsilon^T + \bar{\delta}_\varepsilon^L = \delta\mathbf{I}$ . It is not immediately clear that  $\mathbf{G}^L$  is longitudinal. To prove that indeed it has zero curl, one can apply the unique decomposition of generalized transverse and longitudinal vector fields (as explained in appendix B) to  $\int d\mathbf{r} \mathbf{G}^L \cdot \mathbf{X}$ , where  $\mathbf{X}$  is a general vector field.

The Green tensor  $\mathbf{G}^T$  of the dielectric can be expanded either in terms of the real mode functions  $\mathbf{h}_\lambda$  that were introduced in section 4.2.2, or in terms of complex mode functions  $\mathbf{f}_\lambda$  which are used in chapter 5:

$$\mathbf{G}^T(\mathbf{r}, \mathbf{r}', \omega) = c^2 \sum_\lambda \frac{\mathbf{h}_\lambda(\mathbf{r}) \mathbf{h}_\lambda(\mathbf{r}')}{(\omega + i\eta)^2 - \omega_\lambda^2} = c^2 \sum_\lambda \frac{\mathbf{f}_\lambda(\mathbf{r}) \mathbf{f}_\lambda^*(\mathbf{r}')}{(\omega + i\eta)^2 - \omega_\lambda^2}. \quad (\text{C.4})$$

Both these expansions are equivalent by Eq. (4.23), be it that the real and imaginary parts can be read off more easily in the former form. The term  $i\eta$  in (C.4) makes explicit the positive and infinitesimally small imaginary part of the frequency  $\omega$ . With the positive sign, (C.4) is the causal Green function which transformed back to the time-domain gives a Green function  $\mathbf{G}^T(\mathbf{r}, \mathbf{r}', t - t_0)$  which is nonzero only for positive time differences  $(t - t_0)$ : a cause at time  $t_0$  can only have an effect at later times.

For free space, the transverse and longitudinal delta functions appearing in Eqs. (C.2) and (C.3) are [85]

$$\delta^T(\mathbf{r}) = \frac{2}{3}\delta(\mathbf{r})\mathbf{I} - \frac{1}{4\pi r^3}(\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}) \quad (\text{C.5})$$

$$\delta^L(\mathbf{r}) = \frac{1}{3}\delta(\mathbf{r})\mathbf{I} + \frac{1}{4\pi r^3}(\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}), \quad (\text{C.6})$$

where  $\hat{\mathbf{r}}$  is defined as  $\mathbf{r}/|\mathbf{r}|$ , the unit vector in the direction of  $\mathbf{r}$ . The sum of the transverse and the longitudinal delta function is simply  $\delta(\mathbf{r})\mathbf{I}$ , since their “dipole” parts cancel. The dyadic Green function  $\mathbf{G}_0$  for free space is the sum of a transverse and a longitudinal part. The transverse part is [69]

$$\mathbf{G}_0^T(\mathbf{r}, \omega) = -\frac{\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi(\omega/c)^2 r^3} - \frac{e^{i\omega r/c}}{4\pi r} [P(i\omega r/c)\mathbf{I} + Q(i\omega r/c)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}], \quad (\text{C.7})$$

with the function  $P(z)$  defined as  $\equiv (1 - z^{-1} + z^{-2})$  and  $Q(z)$  as  $(-1 + 3z^{-1} - 3z^{-2})$ . Using the definition (C.3) of the longitudinal Green function and the free-space transverse delta function (C.5), the longitudinal Green function is found to be

$$\mathbf{G}_0^L(\mathbf{r}, \omega) = \frac{\mathbf{I} - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi(\omega/c)^2 r^3} + \frac{\delta(\mathbf{r})}{3(\omega/c)^2}\mathbf{I}. \quad (\text{C.8})$$

The important delta-function term in  $\mathbf{G}_0^L$  appears naturally and there was no need to add it “by hand” as is done elsewhere [4, 69]. Both  $\mathbf{G}_0^T$  and  $\mathbf{G}_0^L$  have nonretarded dipole terms, meaning that a change in a source term changes instantaneously the longitudinal and transverse fields elsewhere. It is only their sum that is fully retarded [85]. The same is true for the Green functions  $\mathbf{G}^T$  and  $\mathbf{G}^L$  of inhomogeneous dielectrics.

In some cases, it is physical to replace the Green functions (C.7) and (C.8) by their angle-averaged values. The angle-averaged value of  $(\hat{\boldsymbol{\mu}} \cdot \hat{\mathbf{r}})^2$  is simply  $\frac{1}{3}$ . Effectively, the averaging amounts to replacing  $\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}$  by  $\frac{1}{3}\mathbf{I}$  both in Eq. (C.7) and in (C.8). The angle-averaged free-space Green function is simply

$$\langle \mathbf{G}_0(\mathbf{r}, \omega) \rangle_{\text{av.}} = -\frac{e^{i\omega r/c}}{6\pi r}\mathbf{I} + \frac{c^2}{3\omega^2}\delta(\mathbf{r})\mathbf{I}. \quad (\text{C.9})$$

In the limit  $\mathbf{r} \rightarrow 0$ , the imaginary part of this angle-averaged Green function still leads to the correct free-space spontaneous-emission rates.

## Appendix D

# Laplace operators and time-dependent coefficients

In section 6.2 the electric-field operator  $\bar{E}(p)$  was given in terms of the operators at  $t = 0$ . Here we give the analogous expressions for the other Laplace operators. Furthermore, we show how to evaluate the time-dependent coefficients  $M_{mn}(t)$  like in Eq. (6.20) for the electric-field operator. Finally, we list the expressions for the coefficients of the other operators.

The expression for  $\bar{E}(p)$  in (6.15) has the following analogous expressions for the other Laplace operators:

$$\begin{aligned}
 \bar{A}(p) &= \frac{1}{\bar{D}(p)} \left\{ -E(0) + pA(0) \right. \\
 &\quad \left. - \frac{\alpha}{\varepsilon_0} \left[ \frac{p^2}{\omega_c^2} \bar{\chi}(p) - 1 \right] X(0) - \frac{1}{\alpha} p \bar{\chi}(p) [P(0) - \bar{B}(p)] \right\}, \\
 \bar{X}(p) &= \frac{1}{\bar{D}(p)} \left\{ -\frac{\varepsilon_0}{\alpha} p \bar{\chi}(p) E(0) + \frac{\varepsilon_0}{\alpha} p^2 \bar{\chi}(p) A(0) \right. \\
 &\quad \left. + \left( \frac{k^2 c^2}{\omega_c^2} + \frac{p^2}{\omega_c^2} + 1 \right) p \bar{\chi}(p) X(0) + \frac{\varepsilon_0}{\alpha^2} (p^2 + k^2 c^2) \bar{\chi}(p) [P(0) - \bar{B}(p)] \right\}, \\
 \bar{P}(p) &= \frac{1}{\bar{D}(p)} \left\{ -\alpha \left[ \frac{p^2}{\omega_c^2} \bar{\chi}(p) - 1 \right] E(0) + \alpha p \left[ \frac{p^2}{\omega_c^2} \bar{\chi}(p) - 1 \right] A(0) \right. \\
 &\quad \left. + \frac{\alpha^2}{\varepsilon_0} \left[ \frac{p^2}{\omega_c^2} \bar{\chi}(p) - 1 \right] \left( \frac{k^2 c^2}{\omega_c^2} + \frac{p^2}{\omega_c^2} + 1 \right) X(0) \right. \\
 &\quad \left. + p \left( \frac{k^2 c^2}{\omega_c^2} + \frac{p^2}{\omega_c^2} + 1 \right) \bar{\chi}(p) [P(0) - \bar{B}(p)] \right\}. \tag{D.1}
 \end{aligned}$$

If we now apply the inverse Laplace transformation to these expressions, we find the full time dependence of the operators  $A$ ,  $X$ , and  $P$ . The inverse Laplace transformation is a contour integration over the Bromwich contour that includes the whole imaginary  $p$

axis. After transforming to frequency variables, the contour includes poles from  $\bar{D}^{-1}(p)$ , which are in the lower half plane and moreover poles on the real frequency axis arising from  $\bar{B}(p)$ . The latter are important in the calculation of the long-time solutions of the operators in section 6.4. However, in the calculation of the coefficients  $M_{mn}(t)$ , which we will discuss here, they play no role. For example, the coefficient  $M_{AE}(t)$  becomes

$$\begin{aligned} M_{AE}(t) &= -\frac{1}{2\pi i} \int_{-i\infty+\eta}^{i\infty+\eta} dp e^{pt} \bar{D}^{-1}(p) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \frac{e^{-i\omega t}}{\varepsilon(\omega)\omega^2 - k^2 c^2} \\ &= \frac{1}{4\pi k c} \int_{-\infty}^{\infty} d\omega \left( \frac{e^{-i\omega t}}{n(\omega)\omega - kc} - \frac{e^{-i\omega t}}{n(\omega)\omega + kc} \right) \\ &= \frac{1}{kc} \sum_j \text{Im}(e^{-i\Omega_j t} \frac{v_{gj}}{c}). \end{aligned} \quad (\text{D.2})$$

Note that  $M_{AE}(t)$  is exponentially damped because all  $\Omega_j$  in the exponentials have negative imaginary parts. The other coefficients can be calculated in a similar way. The results are

$$\begin{aligned} M_{AA}(t) &= M_{EE}(t), \\ M_{AX}(t) &= -\frac{\alpha k}{\omega_c^2 \varepsilon_0 c^4} \sum_j \text{Im} \left[ e^{-i\Omega_j t} v_{gj} \left( 1 - \frac{v_{pj}^2}{c^2} + \frac{\omega_c^2}{k^2 c^2} \right) \right], \\ M_{AP}(t) &= \frac{1}{\alpha} \sum_j \text{Re} \left[ e^{-i\Omega_j t} \frac{v_{gj}}{c} \left( \frac{v_{pj}}{c} - \frac{c}{v_{pj}} \right) \right], \\ M_{XE}(t) &= \varepsilon_0 M_{AP}(t), \\ M_{XA}(t) &= \varepsilon_0 M_{EP}(t), \\ M_{XX}(t) &= -\frac{k^2 c^2}{\omega_c^2} \sum_j \text{Re} \left[ e^{-i\Omega_j t} \frac{v_{gj}}{c} \left( \frac{v_{pj}}{c} - \frac{c}{v_{pj}} \right) \left( 1 - \frac{v_{pj}^2}{c^2} + \frac{\omega_c^2}{k^2 c^2} \right) \right], \\ M_{XP}(t) &= -\frac{\varepsilon_0 k c}{\alpha^2} \sum_j \text{Im} \left[ e^{-i\Omega_j t} \frac{v_{gj}}{c} \left( \frac{v_{pj}}{c} - \frac{c}{v_{pj}} \right)^2 \right], \\ M_{PE}(t) &= \varepsilon_0 M_{AX}(t), \\ M_{PA}(t) &= \varepsilon_0 M_{EX}(t), \\ M_{PX}(t) &= \frac{\alpha^2 k^3 c^3}{\omega_c^4 \varepsilon_0} \sum_j \text{Im} \left[ e^{-i\Omega_j t} \frac{v_{gj}}{c} \left( 1 - \frac{v_{pj}^2}{c^2} + \frac{\omega_c^2}{k^2 c^2} \right)^2 \right], \\ M_{PP}(t) &= M_{XX}(t). \end{aligned} \quad (\text{D.3})$$

With the sum rules discussed in section 6.3, one can see that the “diagonal” coefficients in this list equal 1 at time  $t = 0$ , whereas the other coefficients have the initial value 0.

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# Samenvatting voor iedereen

*De Nederlandse vertaling van de titel van dit proefschrift luidt “Kwantumoptica en meervoudige verstrooiing in diëlektrica”. Om deze titel te verduidelijken worden in de nu volgende samenvatting allereerst enkele relevante begrippen en vraagstellingen besproken. Vervolgens wordt een korte opsomming gegeven van in dit werk gepresenteerde resultaten.*

## Optica en kwantumoptica

### Eigenschappen van licht

Wat is licht? Moeilijk te zeggen, maar veel eigenschappen van licht zijn bekend: dat het zich in rechte lijnen voortbeweegt, mits ongehinderd (zie omslag van dit proefschrift). En dat wit licht uit golven van verschillende kleuren bestaat, elke kleur licht met een eigen golflengte. In een regenwolk of in een prisma worden deze afzonderlijke golven gesplitst. Mensen zien een regenboog met als ene uiterste kleur rood (lange golflengte) en als andere uiterste violet (korte golflengte). De regenboog houdt aan die randen niet op, maar ultraviolette en infrarode golven van hetzelfde (elektromagnetische) type als licht zijn voor mensen onzichtbaar. Een honingbij ziet een andere regenboog dan een mens, met meer kleuren naast het violet.

Licht is een golf en vertoont interferentie, het verschijnsel dat golven elkaar kunnen versterken en uitdoven. Het effect zie je bijvoorbeeld wanneer er een laagje olie op water ligt. Een deel van het licht zal aan de bovenkant van het olielaagje weerkaatsen en een ander deel aan het olie-water oppervlak. Of er sprake is van wederzijdse versterking of uitdoving van deze twee golven hangt sterk af van het verschil in afgelegde afstand en van de golflengte (kleur) van het licht. Een sterk regenboogeffect van olie op water is te verwachten als het olielaagje ongeveer zo dun is als de golflengte van licht, een halve micrometer.

Iets minder in het oog springend is dat licht naast kleur ook polarisatie heeft. Licht is namelijk een transversale golf, wat wil zeggen dat licht dat vooruit gaat, horizontaal of vertikaal kan trillen. Deze twee polarisatierichtingen zijn niet met het blote oog maar wel met polaroidfilters te onderscheiden.

De bovengenoemde eigenschappen van licht zijn vrij bekend en de theorie die dat (en nog veel meer) beschrijft is de klassieke elektrodynamica. Die theorie voldoet in zeer veel gevallen, maar niet als het licht zich als een stroom deeltjes (“fotonen”) voordoet, wat

soms het geval is. Het deeltjeskarakter van licht dringt zich in het dagelijks leven niet op, maar in sommige experimenten wel. Bijvoorbeeld bij het “foto-elektrische effect”: met de energie die in licht zit kun je elektronen losmaken die aan atomen in een vaste stof vastgepind zitten. Het effect treedt alleen op als je op de stof met licht schijnt dat een golflengte heeft kleiner dan een bepaalde waarde. Dit is te verklaren als je aanneemt dat licht-energie in afzonderlijke pakketjes van welbepaalde hoeveelheden (“kwanta”) wordt overgedragen aan materie, waarbij de energie in zo’n pakketje afhangt van de kleur licht.

De moderne theorie die beschrijft hoe licht zowel een golf- als een deeltjeskarakter kan hebben, is de kwantumelektrodynamica. De wisselwerking van zo beschreven licht met materie (atomen) wordt bestudeerd in de kwantumoptica. Dit proefschrift heeft als onderwerp hoe de theorie van de kwantumoptica aangepast moet worden als het licht zich niet in de vrije ruimte maar in een medium bevindt waarin het licht verstrooid of geabsorbeerd kan worden.

### **Atomen als lichtbronnen**

Hoe ontstaat licht? Verreweg het meeste licht om ons heen wordt uitgezonden door atomen, via een proces dat spontane emissie heet. De atomen kunnen zich in verschillende welbepaalde energie-toestanden bevinden, zoals beschreven door de kwantummechanica. Een atoom in een hoog energieniveau zal, mits met rust gelaten, op den duur naar een lagere energietoestand overgaan (“vervallen”). De wet van behoud van energie zegt in dit geval dat het verschil tussen de hoge en de lage energie niet zomaar verdwenen kan zijn. Inderdaad zendt een atoom een lichtdeeltje (“foton”) uit met precies de ontbrekende energie. Het energieverschil van het atoom bepaalt de golflengte en dus ook de kleur van het uitgezonden licht. Elk type atoom heeft zijn eigen energieniveaus. Zodoende zendt elk type atoom zijn eigen kleuren uit. Neem bijvoorbeeld natriumatomen, die als lichtbron worden gebruikt in straatverlichting. Als de lampen net aangaan en nog koud zijn, stralen ze alleen zwakrood licht met lage energie uit, maar even later zijn de lampen heet en vervallen de natriumatomen ook vanaf hogere energieniveaus en schijnen de lampen karakteristiek felgeel. Natuurkundigen kunnen zo aan de hand van gemeten kleuren licht en hun relatieve intensiteit nagaan welk type atomen het licht heeft uitgezonden en bij welke temperatuur; sterrenkundigen leiden op dezelfde manier af welke materie in de ruimte voorkomt en hoe heet sterren zijn.

### **“Spontane” emissie**

Waarom zou een atoom eigenlijk licht uitzenden? Met andere woorden, hoe “weet” een atoom in een hogere toestand dat het naar een lagere toestand kan vervallen onder uitzending van licht? Met deze vraag komen we al dicht bij de inhoud van dit proefschrift. In de klassieke beschrijving van licht en elektromagnetisme is die vraag niet goed te beantwoorden. Volgens de klassieke theorie kan een atoom weliswaar aangezet worden tot het uitzenden van licht onder invloed van al aanwezig licht, maar een enkel atoom in het donker is echt alleen: er is niets dat hem “beweegt” om energie af te geven. Toch stralen atomen in het donker licht uit. “Spontane emissie” is de naam voor een klassiek onverklaarbaar proces.

Volgens de huidige inzichten is spontane emissie geen spontaan proces. Volgens de kwantumelektrodynamica, de opvolger van de klassieke theorie, is er overal om ons heen een elektromagnetisch veld aanwezig. Net als een atoom kan dat veld in verschillende energietoestanden bestaan. Als het veld zich in de laagste energie-toestand bevindt (ook wel de “vacuümtoestand” geheten), is er geen licht. Toch is volgens de kwantumelektrodynamica een atoom in het donker helemaal niet zo alleen: het atoom ziet danwel geen licht, maar voelt wel fluctuaties in het hem omringende elektromagnetische veld. Het zijn de zogenaamde vacuümfluctuaties van het elektromagnetische veld die het atoom aanzetten om zijn licht uit te zenden.

## Levensduur

Er is een typische tijdsschaal waarop het atoom overgaat van het hogere naar het lagere energieniveau onder uitzending van het licht. Deze tijdsschaal heet de “levensduur” van die overgang. Het is niet zo dat een atoom altijd na precies het verstrijken van die levensduur met het licht op de proppen komt. De levensduur is veeleer een gemiddelde tijd waarop licht wordt uitgezonden. De kwantummechanica beschrijft het uitzenden van licht als een statistisch proces en doet met andere woorden slechts uitspraken over de kans dat het atoom binnen een bepaalde tijd vervallen is. Ook de richting waarheen het licht wordt uitgezonden is een kwestie van kansen. Einstein nam precies vanwege dit kans-karakter geen genoegen met de kwantummechanica. Je zou wel een diepere theorie willen hebben die je precies vertelt wanneer een atoom vervalt, maar zo’n theorie is er tot op heden niet (en komt er misschien wel nooit).

## Diëlektrische materialen

### Microscopische en macroscopische beschrijvingen

De golflengte van licht is klein (een halve micrometer, de helft van een duizendste millimeter), maar atomen zijn nog een factor duizend kleiner. Over de afstand van een golflengte van licht zitten in een vaste stof vele honderden atomen op een rij. Door deze schaalverhoudingen is het in de optica mogelijk om veel materialen macroscopisch te beschrijven, in termen van een brekingsindex. “Macroscopisch” duidt erop dat je niet de wisselwerking van licht met alle afzonderlijke atomen beschrijft. De brekingsindex geeft aan hoe de atomen samen het transport van licht vertragen. Glas heeft bijvoorbeeld een brekingsindex van 1.5, wat betekent dat licht in glas anderhalf keer trager gaat dan in de vrije ruimte.

Op soortgelijke manier is de *wave* van publiek bij een sportevenement macroscopisch te beschrijven: die *wave* golft met een snelheid van zo’n twintig meter per seconde meestal tegen de klok in rond het stadion, met een breedte van gemiddeld negen meter (zie [185] en figuur D.1). Individuele mensen komen in deze beschrijving van stadiongolven net zomin voor als individuele atomen bij lichtgolven in vaste stoffen. Er bestaan meer “microscopische” verklaringen voor de snelheid van een stadiongolf aan de hand van reactietijden van voetbalfans; eveneens voor de lichtsnelheid van materie in een vaste stof met behulp van “reactietijden” en polariseerbaarheden van individuele atomen. In dit proefschrift (met uitzondering van hoofdstuk 6) zijn brekingsindices gegeven grootheden.



**Figure D.1:**

De *wave* golft door het stadion (Foto: Universiteit Boedapest).

De macroscopische beschreven materie die een medium vormt voor lichtvoortplanting wordt met de verzamelnaam “diëlektrica” aangeduid. Diëlektrica hebben de eigenschap dat licht zich erin kan voortplanten, ook al wordt het licht soms deels geabsorbeerd. Metalen worden niet tot de diëlektrica gerekend, want in metalen plant licht zich niet eens één enkele golflengte voort.

Een diëlektricum kun je karakteriseren met een brekingsindex die in het algemeen van de plaats en oriëntatie in het medium kan afhangen en verder van alle mogelijke eigenschappen van het licht. Niet alle afhankelijkheden zijn even belangrijk in de praktijk. In dit proefschrift wordt kwantumoptica beschreven in inhomogene diëlektrica (plaatsafhankelijke brekingsindex) en in dispersieve diëlektrica (golflengte-afhankelijke brekingsindex).

### Diëlektrica en spontane emissie

In de vrije ruimte hebben identieke atomen dezelfde levensduren. Je zou daarom denken dat een levensduur een eigenschap van het atoom is. Dat klopt maar voor de helft, want de levensduur hangt evengoed af van de vacuümfluctuaties die het atoom aansporen om licht uit te zenden: in de vrije ruimte is het ook al zo dat licht met kortere golflengte meer fluctuaties voelt en sneller wordt uitgezonden.

De door een atoom gevoelde fluctuaties hangen af van het type diëlektricum waarin het atoom zich bevindt en ook van de specifieke plaats daarbinnen. Voor de mate waarin vacuümfluctuaties een atoom tot vervallen aansporen, kan een handig begrip worden ingevoerd, namelijk de locale toestandsdichtheid: hoe hoger de locale toestandsdichtheid ter plaatse van een atoom, des te sneller het atoom (gemiddeld gesproken) zal vervallen.

Zoals boven vermeld, vertelt het klassieke elektromagnetisme niet waarom atomen vervallen en doet de kwantumelektrodynamica dat wel. Interessant is nu dat de grootte van de locale toestandsdichtheid wel weer met de klassieke elektrodynamica te berekenen is. De “oude” theorie blijft daarvoor dus zeer bruikbaar. De hoofdstukken 2 en 3 lopen op dit in hoofdstuk 5 afgeleide gegeven vooruit en bevatten alleen klassieke elektrodynamica.

### Atomen in de buurt van spiegels

Dat andere materie de levensduur van een atoom beïnvloedt, biedt de mogelijkheid tot “spelen” met die omgeving, om de levensduur veel groter of veel kleiner te maken. Zo is bijvoorbeeld bekend dat dicht bij een spiegel een atoom sneller of langzamer vervalt, al

naar gelang de afstand en de oriëntatie van het atoom ten opzichte van de spiegel. Terwijl een atoom wil vervallen, merkt hij dat zijn spiegelbeeld hetzelfde doet. Dit kan zijn levensduur bespoedigen of vertragen, afhankelijk van de kleur van het uitgezonden licht en de afstand en oriëntatie van het atoom ten opzichte van de spiegel. De typische afstand waarop een spiegel de levensduur van een atoom beïnvloeden kan, is de golflengte van het uitgezonden licht.

### Fotonische kristallen

Een enkel laagje olie op water laat zoals gezegd sommige kleuren licht wel en andere niet door en geeft zo kleurrijke reflecties. Belangrijk daarbij is dat de dikte van het laagje van de orde van de golflengte van licht is. Een sterker kleur-selectief diëlektricum zou je krijgen als meerdere van zulke dunne lagen olie en water elkaar zouden opvolgen. Met vloeistoffen is dat lastig te realiseren, maar opeenvolgende laagjes vaste stoffen met verschillende brekingsindices worden wel gemaakt en heten diëlektrische spiegels. Het zijn een soort ééndimensionale kristallen voor licht. Ze worden gebruikt om licht dat van buiten komt te reflecteren of licht dat er binnenin ontstaat in welbepaalde richtingen naar buiten te laten komen.

Het “spelen” met de omgeving van atomen om hun levensduren en emissie-richtingen te beïnvloeden nam in 1987 een spectaculaire wending toen twee natuurkundigen onafhankelijk van elkaar bedachten dat er inhomogene diëlektrica gemaakt zouden kunnen worden waarbinnen licht van bepaalde energieën helemaal niet zou kunnen bestaan. Het interval van “verboden” energieën heet de “bandkloof” (Engels: “band gap”). Zulke materialen moeten een sterk variërende brekingsindex hebben op een afstandsschaal van de golflengte van licht en worden fotonische kristallen genoemd. Alleen als de variaties van de brekingsindex in drie dimensies voorkomen, dat wil zeggen in lengte, breedte en hoogte, kan er een bandkloof optreden. Bandkloven voor energieën van elektronen waren in de natuurkunde al wel bekend; bandkloven voor energieën van fotonen nog niet.

Een atoom dat in een bandkloof-kristal wordt geplaatst, zal licht met energie in de bandkloof niet kunnen uitzenden. Het elektromagnetische veld kan de energie niet overnemen, omdat die kleur licht eenvoudig niet bestaat in het kristal. Andersom gesteld, er bestaan voor die kleur licht geen vacuümfluctuaties om het atoom aan te sporen dat licht uit te zenden.

Dat spontane emissie tot staan gebracht zou kunnen worden is in de eerste plaats uit fundamenteel oogpunt interessant. In de tweede plaats kan het ook van praktisch belang zijn, omdat spontane emissie in sommige gevallen (in een laser bijvoorbeeld) voor verliezen zorgt. Sinds 1987 is naarstig gezocht naar kristallen met een fotonische bandkloof. De gefabriceerde fotonische kristallen worden steeds beter maar zijn nog niet perfect: een echte fotonische bandkloof bestaat op het moment van schrijven van dit proefschrift nog niet.

## Resultaten

### Spontane emissie in diëlektrische spiegels

In de hoofdstukken 2 and 3 worden diëlektrische spiegels gemodelleerd als oneindig dunne spiegelende vlakken. In een verstrooiingstheorie wordt de T-matrix voor een vlak opgesteld. Voor de T-matrix van een kristal van een eindig aantal vlakken wordt een analytische uitdrukking gevonden, zowel voor scalaire (hoofdstuk 2) als voor vectorgolven (hoofdstuk 3). De Green functie van het eindige kristal van vlakverstrooiers is op een handige manier te bepalen en daarmee de plaatsafhankelijke atomaire levensduren.

### Licht-materie wisselwerking in inhomogene diëlektrica

In hoofdstuk 4 wordt de overgang van het minimale-koppeling formalisme naar het multipoolformalisme uitvoerig behandeld voor microscopisch beschreven gastatomen in macroscopisch beschreven inhomogene diëlektrica. Het vinden van de kanonieke variabelen na keuze van een handige ijk vergt een precieze definitie en berekeningswijze van functionele afgeleides. Een kwantum Hamiltonian wordt gevonden waarin de atomen een multipool-wisselwerking met het elektromagnetische veld hebben maar geen directe wisselwerking met elkaar. Het precieze veld waaraan een atomaire dipool koppelt wordt geïdentificeerd. Op dit niveau van beschrijving blijkt het onontbeerlijk om een locale-veld model te hanteren.

### Kwantumoptica in verstrooiingsperspectief

In hoofdstuk 5 wordt een puntverstrooiersformalisme opgesteld waaruit de electromagnetische veldoperatoren kunnen worden bepaald van een inhomogeen medium met daarin aanwezige gastatomen. Verstrooiing van licht aan gastatomen en spontane emissie van die atomen volgen uit hetzelfde formalisme. Ook meervoudige verstrooiing van kwantumtoestanden van licht kan met dit formalisme worden beschreven.

Er blijkt dat superradiance van naburige atomen onderdrukt kan worden in een inhomogeen medium, vanwege lijnverschuivingen van de afzonderlijke atomen maar ook vanwege verschillen in plaatsafhankelijke levensduren. Zowel de statische als de dynamische dipool-dipoolwisselwerking tussen de gastatomen wordt door het medium gewijzigd.

### Licht in absorberende diëlektrica

Om de commutatierelaties van kanonieke variabelen van een dissipatief system constant te houden in de tijd, is het in de kwantummechanica nodig om het continuüm waarheen de dissipatie plaatsvindt mee te blijven nemen in de theoretische beschrijving. Dat is ook het geval in de kwantumoptica van absorberende diëlektrica. In een reeds bestaand microscopisch model voor een absorberend diëlektricum worden veldoperatoren geïdentificeerd op een manier die handiger is dan via de bekende Fano-diagonalisatie. Door de koppelingssterkte met het continuüm te variëren kunnen verschillende diëlektrische functies gevonden worden in het model. Nabij sterke resonanties van het medium blijkt spontane emissie een tijdsafhankelijk proces.

# Dankwoord

Nu dit proefschrift bijna klaar is, wil ik graag de mensen bedanken die aan de totstandkoming ervan hebben bijgedragen. De meesten van hen werken of werkten aan één van de drie heel verschillende instituten die ik veelvuldig van binnen heb mogen zien: het Van der Waals-Zeeman Instituut (experimentele natuurkunde, Universiteit van Amsterdam), het Instituut voor Theoretische Fysica (ook aan de UvA) en de Faculteit Technische Natuurkunde (nu TNW, Universiteit Twente).

Allereerst wil ik mijn promotor Ad Lagendijk bedanken voor het in mij gestelde vertrouwen, voor de goede begeleiding van mijn promotie-project en voor het samen doen van leuke wetenschap. Ad, vanaf dag één heb je me als de theoreticus aangesproken. Dat petje zakte me eerst wat over de oren, maar ik heb steeds meer mijn draai gevonden. Ik heb je leren kennen als een gedreven wetenschapper en goed manager. Je bent opmerkelijk bereikbaar voor overleg. Aan dat overleg hield ik altijd nieuwe ideeën, een scherpere focus of verse motivatie over. De stoet wetenschappers die je om je heen verzamelt is een groep omdat je gemeenschappelijke belangen formuleert, en blijft hameren op heldere communicatie en op het nemen van eigen verantwoordelijkheid.

Bestaat er *cum laude* voor een co-promotor? Leendert Suttorp, dank voor de vele discussies die we gehad hebben naar aanleiding van mijn berekeningen, zowel voor als na mijn functieverplaatsing naar Twente. Mijn vaak laat aangeleverde berekeningen had je dan toch al weer doorgenomen en dikwijls zelfs met grote precisie nagerekend of uitgebreid. “Leendert doet meer dan van een begeleider mag worden verwacht”, vatte iemand eens samen. Mijn werk ging wekelijks zo door de zeef, waarin gelukkig wel eens wat bleef liggen! En dan vroeg je me nog of ik je terecht als mede-auteur had opgevoerd op een concept-artikel. Dit laat wel zien dat je de lat voor jezelf hoog legt. Een gewone jongen met geldingsdrang moet daar niet altijd aan willen tippen. Naast al het wetenschappelijk-inhoudelijke werk, dank ook voor de vele andere gesprekken en terzijdes.

Toen ik als promovendus begon en nog een eigen klus moest vinden, pingpongede ik mezelf wat teveel tussen bovenstaande zeer verschillende heren. Rudolf Sprik, dank voor procesbegeleiding in die fase. Je kennis is verder verbazend breed. Leuk dat mijn lijnverstrooiers ook in de akoestiek nuttig kunnen zijn, licht en geluid zijn beide golven nietwaar. Willem Vos, je doet mooi onderzoek in de groep Lagendijk-Vos en bent een echte “groupie”: continu bezig mensen met elkaar en met ideeën te koppelen. Dank dat ik een tijdje ergens tussen je bureau en drumstel mocht werken. Gerard Wegdam, je was om een ingewikkelde reden nog even mijn officiële begeleider. Dank voor je enorme inzet

als directeur van het Van der Waals-Zeeman Instituut toen het asbestprobleem acuut werd. Wim Koops, mijn buurman, is een heel bijzondere kerel uut Grunn met wie je over van alles kunt praten, in voor een geintje, belangrijk voor mij en voor de hele Amsterdamse groep (en je schaatste op de Jaap-Edenbaan alle promovendi eruit!).

Tijdens mijn promotietijd heb ik vele groepsleden zien komen en gaan: de omloopsnelheid in de wetenschap is hoog met al die tijdelijke contracten. Johannes de Boer (kwam langs en liet me anderhalf jaar op zijn huis passen in de Pijp), Rik Kop (de enige met een seizoenskaart van Ajax), Mark Kroon (droeg Ad op handen op diens 50<sup>e</sup> verjaardag), Frank Schuurmans (korte broek en Schotse whiskey), Mischa Megens (enthousiasteling, altijd in voor een veganistische mensa-hap in een kraakpand), Henri Schriemer (goed voor stevige discussies), Denis Fenistein (idem), Frank Poelwijk (philosophe chantant), Michiel Thijssen (mijn maatje op Corsica), Anouk Noordervliet, Pedro de Vries ('n echte theoreet, heb veel aan je werk gehad), Judith Wijnhoven (kristallenmaakster van het eerste uur), Duong Hieu Dau, Henk van Driel, Arnout Imhof (puike kamergenoot en gestaag werker), Jaime Gómez-Rivas (met temperament, verschoven bioritme en de nodige aandacht voor de vrijdagmiddagborrels; dank voor je proefschrift-bestanden als voorbeeld), Gijs van Soest (onafhankelijke geest en kenner van het alternatieve film- en muziekcircuit), Yuri Nahmad, Dirk Riese (normaal de rust zelve, maar lyrisch op het toneel), Patrick Johnson (dank voor de goede gesprekken toen we een kamer deelden en voor daarna), Manu Helfer, Juan Lopez Galisteo. Dank voor de goede sfeer.

Dat geldt ook voor de huidige groepsleden: Ivan Nikolaev (kenner van begonia's en gevechtsvliegtuigen), Tijmen Euser (geen mensa meer sinds meneer een baan heeft), Boris Bret (met hart in Portugal en hoofd in Twente, jongleren is er niks bij), Ivo Opstelten (ook in het lab kunstenaar aan het worden), Ari Irman (your English improves and so does your work), Peter Lodahl (inspirerend, nog zo'n liefhebber van kwantum-effecten van licht), Karen van der Molen (net begonnen en nu al assertief), Léon Woldering (met wie het goed loungen is), Cock Harteveld (kent het gebouw en zijn bewoners op zijn duimpje), Raymond Oudeboon (ook voormalig kamergenoot, dank voor het uitzoeken en regelen van van alles en voor je ontregelende humor).

Met Femius Koenderink en Lydia Bechger zat ik in hetzelfde schuitje van proefschrift schrijven en een "dubbelleven" leiden. Leuk dat jullie toch ook meekwamen naar Twente! Lydia, bedankt onder andere voor de zwemlessen en voor het vinden van Surinaams eten hier in 't oosten. Femius onder andere dank voor de cruciale hulp met proefschrift-praktische dingen en voor uitjes naar filmklassiekers. Allebei veel succes gewenst straks. Dat wens ik jou ook, Eliane, en wanneer gaan we lunchen?

De "waterstof-boys" Pepijn Pinkse, Allard Mosk en anderen zijn al een tijdje weg. Allard, leuk dat je nu ook in Twente bent met je aanstekelijke enthousiasme en brede kennis. Dank voor het lezen van delen van dit proefschrift. Met Tom Hijmans en Bert Holsbeke heb ik menige boom opgezet. Jeroen Bongaerts, Joost Peters, Michiel Zwanenburg en Dennis de Lang, we leerden elkaar bij zekere vergaderingen beter kennen. Ronald van Elburg, Mischa Megens, Alejandro Arrizabalaga, Joost Slingerland, en natuurlijk Sander Bais, dank voor de goede samenwerking bij het werkcollege kwantummechanica II.

Voor duizend-en-één dingen kon ik het best bij co-manager Mariet Bos terecht. Secretariële ondersteuning kreeg ik van Ineke Baay, Ina Zwart, Rita Vinig, Paula Uijthoven

en van Karen Munnink. Hartelijk dank allemaal. Bas Benschop bedankt voor de computerondersteuning, het loopt op rolletjes; in Amsterdam zorgden Derk Bouhuijs en Marco Brugmans daarvoor. Mijn werk had inhoudelijk niet veel met technici te maken, maar om sfermakers zoals Flip de Leeuw (ook veel gezien bij CREA), Joost Overtoom (idem achter de tap) en Bert Zwart kun je natuurlijk niet heen. In Enschede geeft Theo Punt me af en toe een kijkje in de Twentse ziel.

I would like to thank all the members of my thesis defense committee for reading the manuscript and for taking part in the defense ceremony.

Annemarieke en Patrick, dank dat jullie mijn paranimfen willen zijn. Kunstenaars Yvonne Grootenboer heeft ook het licht bestudeerd en gaf me spontaan een prachtige dia die nu de kaft van dit proefschrift siert.

Het doen van onderzoek en het schrijven van een proefschrift is met ups en downs gegaan. Echt down was ik toen rugpijn me een tijd dwong thuis te blijven en rust te houden. Vrienden en familie deden inkopen, kookten voor me en beurden me op. Het lijkt gelukkig al lang geleden, maar die hulp vergeet ik niet. Collega's kwamen een gewonnen fles Schotse whiskey met me opdrieken en dat hielp echt.

Een groot deel van mijn vrienden leerde ik kennen bij de studentenvereniging "V.C.S. Hendrik de Cock". Toen samen in een soort dorp, nu verspreid over het (buiten-)land in de meest uiteenlopende bezigheden. De (zeil-)weekenden met "Alles Kabbelt", ik kijk er telkens naar uit. Via medecursisten van het CREA-toneel leerde ik het culturele leven in Amsterdam een beetje kennen. Vrienden, dank voor de steun, maar juist ook voor alle leuke dingen die me van mijn werk afhielden. Tijdens het schrijven van dit boekje was ik niet best bereikbaar. Hoog tijd dat ik velen weer eens spreek.

Broer en vriend David, het is goed reizen met jou. Mijn twee bezoeken aan jou in Bosnië maakten grote indruk op me. Een absoluut hoogtepunt voor mij was jouw huwelijk met Justyna in Nederland én in Polen. Welkom terug allebei vanuit Oslo. Annemarieke, lieve zus, dank voor het attente meeleven met alles wat ik doe. Paul ook bedankt, alleen al voor de vele uitnodigingen voor exposities met jouw mooie werk.

Lieve ouders, dit boekje heb ik aan jullie opgedragen en dat is niet zomaar. Jullie leven al mijn leven lang met me mee en hebben me voor een belangrijk deel gevormd. Onderwijs staat en stond centraal in jullie leven. In het kiezen van een richting hebben jullie me vrijgelaten. Alledrie jullie kinderen hebben met jullie hulp kunnen studeren; en toen ging ik er ook nog Filosofie bij doen. Jullie belangstelling is ook in mijn promotietijd heel belangrijk voor me geweest.

Last but not least, Harmke, dank dat je er bent. Jij hebt van dichtbij mijn werk en de laatste loodjes meegemaakt. Ook je familie leefde mee. Ik was soms humeurig of in mijn hoofd druk met andere dingen bezig en je hebt veel geduld met me gehad (meestal). Je hebt me vaak weer met beide benen op de grond gezet en de zon laten zien, daar ben je zelfs erg goed in. Natuurkunde is je vak niet, maar een aantal opgeblazen zinnen heb je de lezers van dit proefschrift bespaard. Dank voor het maken van de foto op de boekenlegger. Het wordt tijd om samen pootje-badend de golven te bestuderen. Of wordt het "Liggen in de zon"? Ik kijk uit naar de reis die we samen gaan maken!