Quantum optics and multiple scattering in dielectrics
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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 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 $\varepsilon(r)$. A closely related subject of fundamental and also of technological interest is the influence of the dielectric environment on the 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]

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

where $E^{(+)}$ is the positive-frequency part of the electric field operator, and $E^{(-)}$ 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 Atoms as point sources and as point scatterers

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 \( \mathbf{D}(r, t)/[\varepsilon_0 \varepsilon(r)] \) plays an important role, it will be abbreviated in the following as

\[
\mathbf{F}(r, t) = \mathbf{D}(r, t)/[\varepsilon_0 \varepsilon(r)].
\]  (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 \( \varepsilon(r) = 1 \) 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:

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

\[
\mathcal{H} = \mathcal{H}_F + \mathcal{H}_A + \mathcal{H}_{AF}, \quad \text{with}
\]

\[
\mathcal{H}_F = \sum_\lambda \hbar \omega_\lambda a_\lambda^\dagger a_\lambda
\]  (5.4a)

\[
\mathcal{H}_A = \sum_{m=1}^N \hbar \Omega_m b_m^\dagger b_m
\]  (5.4b)

\[
\mathcal{H}_{AF} = - \sum_{m=1}^N \mu_m \cdot \mathbf{F}(\mathbf{R}_m) = \sum_{m, \lambda} (b_m + b_m^\dagger)(g_{\lambda m} a_\lambda + g^{*}_{\lambda m} a_\lambda^\dagger).
\]  (5.4c)

The atomic frequencies \( \Omega_m \) and transition dipole moments \( \mu_m \) 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 \( \mu_m \) 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

$$g_{\lambda m} = -i \sqrt{\frac{\hbar \omega_\lambda}{2\varepsilon_0}} \mu_m : f_\lambda(R_m).$$

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:

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

$$\dot{a}_\lambda^\dagger(t) = i \omega_\lambda a_\lambda^\dagger(t) + \frac{i}{\hbar} \sum_m g_{\lambda m} \left[ b_m(t) + b_m^\dagger(t) \right].$$

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

$$\dot{b}_m(t) = -i \Omega_m b_m(t) - \frac{i}{\hbar} \sum_\lambda \left[ g_{\lambda m} a_\lambda(t) + g_{\lambda m}^* a_\lambda^\dagger(t) \right]$$

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

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

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

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 $a_\lambda^{(0)} + i \omega_\lambda a_\lambda^{(0)} = 0$. After partially integrating, the Laplace transform of this equation becomes

$$-i(\omega - \omega_\lambda) a_\lambda^{(0)}(\omega) = a_\lambda^{(0)}(t = 0).$$
Also in Laplace language, the equations for the atomic operators become

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

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

\[ a_\lambda(\omega) = a_\lambda^{(0)}(\omega) + \frac{i\hbar^{-1}}{\omega - \omega_\lambda} \sum_m g_{\lambda m}^* \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \left\{ g_{\lambda' m} a_{\lambda' m}(\omega) + g_{\lambda' m}^* a_{\lambda' m}^\dagger(\omega) \right\}. \]  
(5.11a)

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

\[ a_\lambda^\dagger(\omega) = a_\lambda^{(0)\dagger}(\omega) - \frac{i\hbar^{-1}}{\omega + \omega_\lambda} \sum_m g_{\lambda m} \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \left\{ g_{\lambda' m} a_{\lambda' m}(\omega) + g_{\lambda' m}^* a_{\lambda' m}^\dagger(\omega) \right\}. \]  
(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)

\[ F(\mathbf{r}, \omega) = E^{(0)}(\mathbf{r}, \omega) \]
\[ + \sum_m K(\mathbf{r}, \mathbf{R}_m, \omega) \cdot S_m(\omega) + \sum_m K(\mathbf{r}, \mathbf{R}_m, \omega) \cdot V_m(\omega) \cdot F(\mathbf{R}_m, \omega). \]

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 $E^{(0)}(r, \omega)$ is the electric field in the absence of the atoms, with both the $(\pm)$ and the $(\mp)$ parts; the atomic source operators $S_m(\omega)$ are vectors and have the form

$$S_m(\omega) \equiv \hat{\mu}_m S_m(\omega) \equiv \hat{\mu}_m \left( -\frac{i\mu_m \omega^2}{\varepsilon_0 c^2} \right) \left[ \frac{b_m(0)}{\omega - \Omega_m} + \frac{b_m^+(0)}{\omega + \Omega_m} \right].$$  

(5.13)

where $\hat{\mu}_m$ denotes the unit vector in the direction of the atomic dipole moment $\mu_m$. Notice that $S_m$ features the atomic creation and annihilation operators at the initial time zero: in quantum optics, the atomic variables cannot 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

$$V_m(\omega) \equiv \hat{\mu}_m V_m(\omega) \hat{\mu}_m \equiv \hat{\mu}_m \left( \frac{\mu_m^2 \omega^2}{h\varepsilon_0 c^2} \right) \left( \frac{2\Omega_m}{\omega^2 - \Omega_m^2} \right) \hat{\mu}_m.$$  

(5.14)

The potentials $V_m(\omega)$ are tensorial instead of isotropic because the atoms cannot 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

$$\alpha_{BM}(\omega) = \frac{2\mu_m^2 \Omega_m}{h\varepsilon_0 (\Omega_m^2 - \omega^2)}.$$  

(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 $K$ which is given by

$$K(r, r', \omega) \equiv c^2 \sum_\lambda \frac{f_{\lambda}(r) f_{\lambda}^*(r') \omega_{\lambda}^2}{(\omega^2 - \omega_{\lambda}^2) \omega^2}.$$  

(5.16)

In the denominator, the frequency $\omega$ is assumed to contain an infinitesimally small positive imaginary part $i\eta$, so that $K$ has an imaginary part whenever $\omega$ coincides with a mode frequency $\omega_{\lambda}$. The dyadic $K$ 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 $G(r, r', \omega)$ of the inhomogeneous dielectric is the solution of the wave equation

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

(5.17)

where the right-hand side is the ordinary Dirac delta function times the unit tensor. The generalized transverse Green function $G_T$ is the solution of

$$-\nabla \times \nabla \times G_T(r, r', \omega) + \varepsilon(r)(\omega/c)^2 G_T(r, r', \omega) = \delta_T(r', r).$$

(5.18)
The source term $\delta^T_\varepsilon$ 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 $G^T$ has an expansion

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

(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 $K$ (5.16) can be rewritten as

$$K(r, r', \omega) = \frac{c^2}{\omega^2 - \omega_\lambda^2} \sum_{\lambda} f_{\lambda}(r) f^*_\lambda(r') - \frac{1}{(\omega/c)^2 \varepsilon(r')} \delta^T_\varepsilon(r, r').$$

(5.20)

It consists of the generalized transverse Green function (5.19) and a term proportional to the generalized transverse delta function $\delta^T_\varepsilon$ (4.24). Both terms are medium-dependent. Note that $K$ is generalized transverse in its variable $r$. In particular, the second term in (5.21) is not a longitudinal term. Because of its transversality property, $K$ 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 $K$ as

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

(5.22)

According to this identity, the dyadic $K$ 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 $r$ and $r'$ 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 $-\mu \cdot D/\varepsilon \varepsilon_0 (R)$ interaction that was derived in the previous chapter. Although it is different from $G$, the quantity $K$ 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 $F(r, \omega)$ 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):

$$A(r, \omega) = \sum_{\lambda} \sqrt{\frac{\hbar}{2 \varepsilon_0 \omega_\lambda}} \left[ a_\lambda(\omega) f_\lambda(r) + a^*_\lambda(\omega) f^*_\lambda(r) \right]$$

(5.23)

$$= A^{(0)}(r, \omega) + \frac{1}{i \omega} \sum_m G^T(r, R_m, \omega) \cdot [S_m(\omega) + V_m(\omega) \cdot F(R_m, \omega)].$$

Here the vector potential is propagated by the generalized transverse Green function $G^T$, rather than by $K$, 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

\[
\mathbf{P}_{gm}(r,t) = \delta(r - \mathbf{R}_m) \mathbf{P}_{gm}(t) = \delta(r - \mathbf{R}_m) \mu_m \left[ b_m(t) + b^*_m(t) \right].
\]  \tag{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

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

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

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

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
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 $\mu$ 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 $K$ in that equation cannot be replaced by the Green function $G$, since the delta function in $K$ 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

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

In this equation, $K$ and $V$ are classical quantities, whereas $F$, $E^{(0)}$, and $S$ 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:

\[ F(r, \omega) = F_1(r, \omega) + F_2(r, \omega), \quad \text{where} \]
\[ F_1(r, \omega) = E^{(0)}(r, \omega) + K(r, R, \omega) \cdot T(\omega) \cdot E^{(0)}(R, \omega) \quad (5.31) \]
\[ F_2(r, \omega) = K(r, R, \omega) \cdot S(\omega) + K(r, R, \omega) \cdot T(\omega) \cdot K(R, R, \omega) \cdot S(\omega). \quad (5.32) \]

In this exact solution for the Field, $F_1$ describes light that is either undisturbed or scattered by the guest atoms; the source Field $F_2$ can be related to spontaneous emission by the atoms. The solutions of $F_2$ and $F_1$ will be discussed here and in section 5.3.2, respectively.

The single-atom T-matrix is

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

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

\[ F_2(r, \omega) = \frac{K(r, R, \omega) \cdot S(\omega)}{1 - \hat{\mu} \cdot K(R, R, \omega) \cdot \hat{\mu} V(\omega)}, \quad (5.34) \]

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

\[ F_2(r, t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \ e^{-i\omega t} \frac{(\omega^2 - \Omega^2)K(r, R, \omega) \cdot S(\omega)}{\omega^2 - \Omega^2 - \hat{\mu} \cdot K(R, R, \omega) \cdot \hat{\mu}[2\mu^2\omega^2\Omega/(\hbar c_0 c^2)]}. \quad (5.35) \]

This integral in general cannot be evaluated further without the explicit knowledge of the Green function. Light can go from the source at $R$ to a detector at position $r$ 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 $r$. 
One can only say something about the the amplitude at position \( r \) 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

\[
\Gamma/2 = -\mu \cdot \text{Im} \left[ \mathbf{K}(\mathbf{R}, \mathbf{R}, \Omega) \cdot \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 r^2} \right].
\]  

(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 \( \mathbf{G}^\parallel \) (C.3) is real, the decay rate is proportional to the imaginary part of only \( \mathbf{G}^\parallel \). 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 \( \mathbf{G}^\parallel_0(\mathbf{R}, \mathbf{R}, \Omega) \) is equal to \(-\Omega/(6\pi\varepsilon_0)\) [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

\[
\Delta' = \hat{\mu} \cdot \text{Re} \left[ \mathbf{K}(\mathbf{R}, \mathbf{R}, \Omega) \cdot \hat{\mu} \left( \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 r^2} \right) \right].
\]  

(5.37)

For two reasons, this frequency shift is infinitely large even in free space. The first reason is that the delta function term \( \delta(\mathbf{r} - \mathbf{R}_m)I/\epsilon(\mathbf{r})\omega_0^2 \) 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 \( \epsilon(\mathbf{r}) \). 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 \( \omega_c = 2\pi c/a \), 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

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

(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 $K$ in the denominator of the expression (5.33) for the single-atom T-matrix must be replaced by $G(R, R, \omega) - \text{Re} \, G_0(R, R, \omega)$.

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 $F_2$. There is also the other contribution $F_1$ to the Field, consisting of the undisturbed electric field and a scattered field:

$$F_1(r, \omega) = E^{(0)}(r, \omega) + K(r, R, \omega) \cdot T(\omega) \cdot E^{(0)}(R, \omega).$$

(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 $F_1$ 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 $F_1$ featuring the annihilation operators, one finds

$$F_1^{(+)}(r, t) = E^{(0)(+)}(r, t) + \sum_\lambda \frac{\mu^2}{2\pi \hbar c^2} \sqrt{\frac{\hbar \omega_\lambda}{2\varepsilon_0}} a^{(0)}(0) f_\lambda(r) \cdot \hat{\mu} \times$$

$$\int_{-\infty}^{\infty} d\omega e^{-i\omega t} \left\{ \begin{array}{l}
\frac{2\Omega_\omega \omega_\lambda^2 K(r, R, \Omega_s) \cdot \hat{\mu}}{\Omega_s^2 - \omega_\lambda^2 + \Gamma^2/4 - i\omega_\lambda \Gamma} \cdot \frac{1}{\omega - \omega_\lambda} \\
+ \frac{\Omega_\omega^2 K(r, R, \Omega_s) \cdot \hat{\mu}}{\Omega_s - \omega_\lambda - i\Gamma/2} \cdot \frac{1}{\omega - \Omega_s + i\Gamma/2} \\
+ \frac{\Omega_\omega^2 K'(r, R, \Omega_s) \cdot \hat{\mu}}{\Omega_s + \omega_\lambda + i\Gamma/2} \cdot \frac{1}{\omega + \Omega_s + i\Gamma/2} \end{array} \right\}$$

(5.40)

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 $F^{(-)}$ of the Field equals $[F^{(+)}]^\dagger$. 

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Point scatterers and quantum optics in inhomogeneous dielectrics
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 $F_1$.

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 $R = 0$ is assumed; $\langle I_s \rangle$ and $\langle I_p \rangle$ are the expectation values of the scattered intensity at position $r$ and of the pump intensity, respectively [15]. Assume monochromatic polarized pump light with wave vector $k_p$, frequency $\omega_p = k_p c$ and polarisation direction $e_p$ in a coherent state $\langle \alpha_p \rangle$. The pump light intensity is $\hbar \omega_p |\alpha_p|^2$, where Eq. (5.1) was used for the intensity operator. The average intensity of the scattered light (with polarisation direction $e_s$) 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

$$\frac{d\sigma}{d\Omega} = \frac{\langle \mu \cdot e_p \rangle^2 \langle \mu \cdot e_s \rangle^2}{4\pi^2 \hbar^2 \varepsilon_0^2 c^4} \cdot \frac{\Omega_s^2 \omega_p^4}{(\Omega_s^2 - \omega_p^2 + \Gamma^2/4)^2 + \omega_p^2 \Gamma^2}. \tag{5.41}$$

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 $\Gamma(R, \Omega)$ (5.36) and line shifts $\Delta(R, \Omega)$ (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 \( \varepsilon(r) = \varepsilon = n^2 \). 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 \( G_{\text{macro}} \) be the Green function for the homogeneous medium with \( \varepsilon \) a constant everywhere. Compare the wave equation (C.1) for this Green function when \( \varepsilon(r) = \varepsilon \) with the free-space wave equation for \( G_0 \) with \( \varepsilon(r) = 1 \). It follows that \( G_{\text{macro}}(r,\omega) \) is equal to \( G_0(r,n\omega) \). This identity also holds for the transversal (C.7) and longitudinal (C.8) parts separately:

\[
G_{\text{macro}}^T(r,\omega) = -\frac{1-3\hat{r} \otimes \hat{r}}{4\pi(n\omega/c)^2 r^3} e^{i n\omega r/c} \mathbf{I} + Q(i n\omega r/c) \hat{r} \otimes \hat{r} \\
G_{\text{macro}}^L(r,\omega) = \frac{1-3\hat{r} \otimes \hat{r}}{4\pi(n\omega/c)^2 r^3} + \frac{\delta(r)}{3(n\omega/c)^2} \mathbf{I},
\]

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 \( \omega_k = kc/n \). 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 \( \omega_k = kc/n \) 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 \( [\varepsilon(r) - 1] \) 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-
5.3 Single-atom properties altered by the medium

\[ \mathbf{V}_{\sigma} \equiv \theta(R - |r|)(\varepsilon - 1)(\omega/c)^2 \mathbf{l}. \] (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 \( f_{k,\sigma}^{\text{macro}}(r) \) is equal to \( \sigma_k \exp(ik \cdot r)/\sqrt{\varepsilon} \).

The guest atom couples to the Field at \( \mathbf{r} = 0 \). In the origin, the mode function \( f_{k,\sigma}^{\text{c}} \) of the dielectric-with-hole satisfies the Lippmann-Schwinger equation

\[ f_{k,\sigma}^{\text{c}}(0) = \sigma_k + (\varepsilon - 1)(\omega/c)^2 \int_{\Omega} \, dr' \, G_{\text{macro}}(-r', \omega) \cdot f_{k,\sigma}^{\text{c}}(r'). \] (5.44)

The Green function in this equation is the sum of the transverse and longitudinal Green functions (5.42). Now since \( k_R < 1 \) is assumed, the short-distance behavior of the Green function \( G_{\text{macro}} \) is important. In that limit, \( G^T \) has an integrable divergence, as follows from Eq. (5.42). The integral over the cavity volume of \( G^T \) 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 \( g_0(r) = -\exp(i\omega \mathbf{r})/(4\pi r) \) is integrable as well.) On the other hand, Eq. (5.42) also shows that \( G^L \) 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 \( G^L \) 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

\[ f_{k,\sigma}^{\text{c}}(0) = \left( \frac{3\varepsilon}{2\varepsilon + 1} \right) f_{k,\sigma}^{\text{macro}}(0). \] (5.45)

The factor \( L_c \equiv 3\varepsilon/(2\varepsilon + 1) \) 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 \( G^c(0, 0) \) is enhanced by a factor \( L_c^2 \) as compared to \( G_{\text{macro}}(R, R) \); the spontaneous-emission rates should be enhanced by \( L_c^2 \) 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 \( f_{\sigma} \) must be used rather than the plane-wave modes \( f_{\text{macro}} \). 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 \( f_{\sigma}(\mathbf{r}) \) and \( f_{\sigma}^{\text{macro}}(\mathbf{r}) \) as described in Eq. (5.44) can be neglected for positions \( \mathbf{r} \neq \mathbf{R} \). Consequently, one can say that

\[ \mathbf{G}_{\sigma}(\mathbf{r}, \mathbf{r}', \omega) = [L_c]^{2n} \mathbf{G}_{\text{macro}}(\mathbf{r}, \mathbf{r}', \omega). \] (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 \( F_1 \) and \( F_2 \). The scattered Field \( F_1 \) solves the homogeneous and the source field \( F_2 \) the inhomogeneous wave equation:

\[
F_1(r, \omega) = E^{(0)}(r, \omega) + \sum_{m,n=1}^{N} K(r, R_m, \omega) \cdot T_m^{(N)}(\omega) \cdot E^{(0)}(R_n, \omega).
\]  

(5.47)

The \( N \)-atom T-matrix can indeed be calculated explicitly and has the form

\[
T_m^{(N)}(\omega) = \tilde{\mu}_m T_m^{(N)}(\omega) \tilde{\mu}_n = \tilde{\mu}_m T_m(\omega) M_m^{(-1)}(\omega) \tilde{\mu}_n.
\]  

(5.48)

where the \( N \times N \) matrix \( M(\omega) \) is defined as

\[
M_{ij}(\omega) = [\delta_{ij} - (1 - \delta_{ij})\tilde{\mu}_i \cdot G(R_i, R_j, \omega) \cdot \tilde{\mu}_j].
\]  

(5.49)

The basic ingredients of this T-matrix are the Green function \( G \) 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 \( \tilde{\mu}_m \cdot G(R_m, R_n, \omega) \cdot \tilde{\mu}_n \) can be determined. In the matrix \( M \) the Green function \( K(5.22) \) should have shown up instead of the \( G \), 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 \( G \) anymore. Equations (5.48) and (5.49) neatly sum up infinitely many scattering events which are not described by \( G \). The occurrence of \( G \) in Eq. (5.49) signifies that light does propagate as described by \( G \) 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 \( G_0 \), respectively.

By using the same iteration that led to the expression (5.47) for \( F_1 \), the source-field \( F_2 \) can also be expressed in terms of the T-matrix:

\[
F_2(r, \omega) = \sum_{m=1}^{N} K^{(N)}(r, R_m, \omega) \cdot S_m(\omega).
\]  

(5.50)
Here, $K^{(N)}$ is the Green function of the inhomogeneous dielectric including the $N$ atoms, also including self-interactions of the atoms:

$$K^{(N)}(\mathbf{r}, \mathbf{r}', \omega) = K(\mathbf{r}, \mathbf{r}', \omega) + \sum_{m,n=1}^{N} K(\mathbf{r}, \mathbf{R}_m, \omega) \cdot \mathbf{T}^{(N)}_{mn}(\omega) \cdot K(\mathbf{R}_n, \mathbf{r}', \omega). \quad (5.51)$$

At positions $\mathbf{r}$ and $\mathbf{r}'$ different from one of the guest atoms, $K^{(N)}(\mathbf{r}, \mathbf{r}', \omega)$ is equal to what one would call the total Green function $G^{(N)}$ of both the dielectric and its guests. The solution (5.50) shows that the source-field $K^{(N)}(\mathbf{r}, \mathbf{R}_m, \omega) \cdot S_m(\omega)$ 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 $S_p$:

$$S_p(\mathbf{r}, t) = \delta(\mathbf{r} - \mathbf{r}_p) \left[ s_p e^{-i\omega_p t} + s_p^* e^{+i\omega_p t} \right]. \quad (5.52)$$

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

$$\mathbf{E}(\mathbf{r}, t) = \int d\mathbf{r}' \int_0^t dt' K^{(N)}(\mathbf{r}, \mathbf{r}', t - t') \cdot S_p(\mathbf{r}', t')$$

$$= \frac{1}{2\pi i} \int_{-\infty}^{\infty} d\omega e^{-i\omega t} K^{(N)}(\mathbf{r}, \mathbf{r}_p, \omega) \cdot \left[ \frac{s_p}{\omega - \omega_p} + \frac{s_p^*}{\omega + \omega_p} \right]. \quad (5.53)$$

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 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 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 Ba$^+$ ions that emit at a wavelength of 493 nm and for well-defined separations $|\mathbf{R}|$ around 1.5 $\mu$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 Two-atom superradiance in inhomogeneous medium

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:

\[
F_2(r, \omega) = K^{(2)}(r, R_1, \omega) \cdot S_1(\omega) + K^{(2)}(r, R_2, \omega) \cdot S_2(\omega). \tag{5.54}
\]

The goal is now to calculate the Green function \( K^{(2)} \) 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 \( K^{(2)} \) 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 \( K \) of the dielectric in (5.22). The calculation therefore starts with the matrix

\[
M^{(2)} = \begin{pmatrix}
1 & -\hat{\mu}_1 \cdot G(R_1, R_2, \omega) \cdot \hat{\mu}_2 T_2 \\
-\hat{\mu}_2 \cdot G(R_2, R_1, \omega) \cdot \hat{\mu}_1 T_1 & 1
\end{pmatrix}. \tag{5.55}
\]

Let \( J_{12} \) be short-hand notation for \( \beta \mu_1 \cdot G(R_1, R_2, \omega) \cdot \hat{\mu}_2 \), with \( \beta \) defined as \( \mu^2 \omega^2/(\hbar^2 \varepsilon_0 c^2) \). Then \( J_{21} = J_{12} \) and the two-atom T-matrix is

\[
T^{(2)} = \frac{1}{1 - T_{11} J_{12}^2 T_2 / \beta^2} \begin{pmatrix}
\mu_1 \mu_1 T_1 & \mu_1 \mu_2 T_2 J_{12} T_2 / \beta \\
\mu_2 \mu_1 T_2 J_{12} T_1 / \beta & \mu_2 \mu_2 T_2
\end{pmatrix}. \tag{5.56}
\]

Each of the four matrix elements of \( T^{(2)} \) is a dyadic of the same type as the single-atom T-matrix (5.33). The frequency dependence in the Green function \( K(r, R_1, \omega) \) will be dropped and it will be further abbreviated as \( K(r_1) \); other terms are written in similar short-hand notation. The Green function \( K^{(2)}(r_1) \) can be written with Eq. (5.51) as

\[
K^{(2)}(r_1) = K(r_1) \cdot \left[ 1 + T_{11}^{(2)} \cdot K(11) + T_{12}^{(2)} \cdot K(21) \right] \\
+ K(r_2) \cdot \left[ T_{21}^{(2)} \cdot K(11) + T_{22}^{(2)} \cdot K(21) \right]. \tag{5.57}
\]

It is convenient to split off the vector character from the source terms \( S_j \) (5.13) by rewriting them as \( \hat{\mu}_j S_j \). Use Eq. (5.56) to rewrite the T-matrix elements of \( T^{(2)} \) 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

\[
K^{(2)}(r_1) \cdot S_1 = \left( \frac{1 + T_1 X_1 / \beta}{1 - T_{11}^2 T_2^2 / \beta^2} \right) \left[ K(r_1) \cdot \hat{\mu}_1 + K(r_2) \cdot \hat{\mu}_2 T_2 J_{21} / \beta \right] S_1. \tag{5.58}
\]

Here, \( X_1 \) is defined as

\[
X_1 = \Omega_1(\omega) - \Omega = \Delta_1(\omega) - i \Gamma_1(\omega)/2. \tag{5.59}
\]
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

$$K^{(2)}(r_1) \cdot S_1 = \frac{(\omega^2 - \Omega^2)S_1 \left[ K(r_1) \cdot \mu_1 \left( (\omega^2 - \Omega^2) - 2\Omega X_2 \right) + K(r_2) \cdot \mu_2 2\Omega J_{12} \right]}{(\omega^2 - \Omega^2)^2 - 2\Omega (X_1 + X_2) (\omega^2 - \Omega^2) + 4\Omega^2 (X_1 X_2 - J_{12}^2)}. \tag{5.60}$$

The denominator carries the important information about the resonance frequencies $\Omega_{\pm}(\omega)$ of the two-atom system:

$$\Omega_{\pm}^2 = \Omega^2 + 2\Omega \left[ \frac{1}{2} (X_1 + X_2) \pm \sqrt{\left( \frac{X_1 - X_2}{2} \right)^2 + J_{12}^2} \right]\approx \Omega_1 + \Omega_2 \pm \sqrt{\left( \frac{\Omega_1 - \Omega_2}{2} \right)^2 + J_{12}^2}. \tag{5.61}$$

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 $S_1$, one has

$$K^{(2)}(r_1) \cdot S_1 = \sum_{\pm} \left( \frac{-i\mu \omega^2}{4\varepsilon_0 c^2 \Omega_{\pm}} \right) [K(r_1) \cdot \mu_1 (1 \pm \sin \alpha) \pm K(r_2) \cdot \mu_2 \cos \alpha] \times \left[ (\omega + \Omega) b_1(0) + (\omega - \Omega) b_1^*(0) \right] \left[ \frac{1}{\omega - \Omega_{\pm}} - \frac{1}{\omega + \Omega_{\pm}} \right]. \tag{5.62}$$

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:

$$\sin \alpha \equiv \frac{(\Omega_1 - \Omega_2)}{\sqrt{(\Omega_1 - \Omega_2)^2 + 4J_{12}^2}}; \quad \cos \alpha \equiv \frac{2J_{12}}{\sqrt{(\Omega_1 - \Omega_2)^2 + 4J_{12}^2}}. \tag{5.63}$$

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 $F_2(r, t)$ 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:

$$F_2(r, t) = \sum_\pm \{ L_{1\pm}(r, t) b_1(0) + L_{2\pm}(r, t) b_2(0) + \text{H.c.} \}. \quad (5.64)$$

The vectors $L_{1\pm}$ are defined as

$$L_{1\pm}(r, t) \equiv \frac{-i \mu \Omega_\pm^2}{2 \varepsilon_0 c^2} \int_{-\infty}^{\infty} d\omega \frac{e^{i\omega t}}{\omega - \Omega_\pm} \times \left[ K(r, R_1, \Omega_\pm) \cdot \mu_1 (1 \pm \sin \alpha_\pm) \pm K(r, R_2, \Omega_\pm) \cdot \mu_2 \cos \alpha_\pm \right]. \quad (5.65)$$

where $\alpha_\pm$ is written as short-hand notation for $\alpha(\Omega_\pm)$. The vectors $L_{2\pm}$ 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

$$\langle I(r, t) \rangle = 2 \varepsilon_0 c | L_{1+}(r, t) + L_{1-}(r, t) |^2. \quad (5.66)$$

The time-dependent intensity that passes at $r$ is a complicated interference pattern of light coming from four sources, a fast(er) and a (more) slowly decaying source at $R_1$, and also a fast and a slow source at the atomic position $R_2$. 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 $\Delta_{12}$ 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 $X_1 = X_2 = X^{(0)}$ 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 $|R_1 - R_2| \rightarrow 0$: 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 $F_2^{2nd}(r, t)$ of the total source field $F_2(r, t)$ that is finally emitted from the initially unexcited second atom. This part consists of a faster and a slower source term:

$$F_2^{2nd}(r, t) = -\frac{i\mu b_1(0)}{2\varepsilon_0 c^2} \sum_{\pm} \int d\omega \frac{\pm \omega^2 \cos \alpha_\pm \mathbf{G}(r, \mathbf{R}_2, \Omega_\pm) \cdot \mathbf{G} \cdot \mathbf{\mu}_2 e^{-i\omega t}}{\omega - \Omega_\pm} + \text{H.c.} \quad (5.67)$$

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 $\varepsilon(r) = \varepsilon = n^2$. The results of two-atom superradiance of
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 \( \mu_{1,2} \) are equal in magnitudes \( \mu \) and orientations and assume further that \( \hat{\mu}_{1,2} \cdot \mathbf{R} = 0 \) ("\( \Sigma \)-configuration"). With Eq. (5.46) one finds the dipole-dipole interaction

\[
J_{12}(n, R, \Omega) = -\left(\frac{\mu^2 \Omega^2}{\hbar \epsilon_0 c^2}\right) \frac{e^{i n \Omega R/c}}{4\pi R} \left\{ 1 + \frac{ic}{n\Omega R} - \frac{c^2}{n^2 \Omega^2 R^2} \right\} \left( \frac{3n^2}{2n^2 + 1} \right)^2. \tag{5.68}
\]

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 \( L^2_{\text{e}} \) 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 \( L^2_{\text{e}} \). 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

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**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: \( n = 3.0 \).
for the dipole orientations chosen. For two guest atoms in a gas, relative orientations $\mathbf{R}$ are not fixed and the average interaction is described by the angle-averaged Green function $\mathcal{L}_c \langle \mathbf{G}_0(\mathbf{R}, n\Omega) \rangle_{\text{av}}$, 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

$$J_{12}(n, \mathbf{R}) = \left(\frac{\mu^2}{4\pi \hbar c_0 R^3}\right) \frac{1}{n^2} \left(\frac{3n^2}{2n^2 + 1}\right)^2.$$  (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 these 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 $n\mathcal{L}_c^2\Gamma_0$. 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
5.7 Discussion: superradiance in photonic crystals

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 \( \Delta(R_{1,2}) \) (5.38) and on the decay rates \( \Gamma(R_{1,2}) \) (5.36):

\[
\Omega_1 - \Omega_2 = \Delta(R_1) - \Delta(R_2) - i \left[ \Gamma(R_1) - \Gamma(R_1) \right] / 2. \tag{5.70}
\]

Superradiance will only occur when \(|\Omega_1 - \Omega_2|/2\) is smaller than the absolute value of the dipole-dipole interaction

\[
J_{12}(R_1, R_2, \Omega) = \frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \mu_1 \left\{ c^2 \sum_{\lambda} \frac{f_{\lambda}(R_1) f_{\lambda}^*(R_2)}{\Omega^2 - \omega_{\lambda}^2} - c^2 \sum_{\lambda} \frac{f_{\lambda}(R_1) f_{\lambda}^*(R_2)}{\Omega^2} \right\} \mu_2, \tag{5.71}
\]

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 \( \mathbf{G}^F(R_1, R_2, \Omega) \) (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 \( \mathbf{G}^L(R_1, R_2, \Omega) \) (C.3). This second term is the “static dipole-dipole interaction”, or SDDI. Both the resonant and the static dipole-dipole interactions are be 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 $k\epsilon = n\omega k$ also for these large wave vectors and corresponding high frequencies. In reality, $\nu(\omega)$ 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 λ, 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.