Spontaneous emission near resonant optical antennas

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Scattering and Spontaneous Emission by Electric Dipoles

This Chapter gives an introduction to the theoretical framework that is used to describe the scattering of light by small particles throughout this thesis. Furthermore, it describes the classical electrodynamic approach taken to calculate the decay-rate enhancement of dipolar emitters in complex photonic environments. This Chapter does not contain any new results that could not be found in the literature. It is much rather intended as a concise introduction to dipolar scattering theory for the unfamiliar reader in order to make the theoretical part of this thesis mostly self contained.

4.1 Introduction

The theory of electrodynamics might seem simple when considering that it is set entirely by the four Maxwell equations together with two constitutive equations describing the involved materials [1]. It however turns out that when considering any but the simplest electrodynamic problem, finding a solution to Maxwell's equations is challenging to say the least. Therefore, a variety of schemes and techniques have been developed in order to simplify the search for valid solutions [2]. One well known example is the concept of lumped circuit elements. In a lumped-element framework anyone mastering basic arithmetics can understand and design electrical circuits with powerful functionality and therefore put Maxwell's equations to work [3]—under the constraint that the circuit

formalism describing resistors, capacitors, and inductors connected with conductors breaks down when the signal frequencies correspond to wavelengths that become comparable to the physical circuit size or electrodynamic effects like radiation become important [4].

In this Chapter we discuss another simplification, namely the dipolar approximation leading to a coupled-dipole model [5]. In optics, point-dipole methods are an approximation toolbox as powerful as lumped circuits are in the electrostatic regime [2]. It is its electrodynamic nature that makes a point-dipole model superior to lumped-element descriptions of photonic circuits at optical frequencies [6–8].

The dipolar approximation is a good description of a scatterer if the currents oscillating at frequency ω and thereby generating the fields are constrained to a volume $d^3 \ll \lambda^3$, where $\lambda = 2\pi c/\omega$. Furthermore, the observer, or any other current distribution (i.e. scatterer), must be sufficiently far away, such that $d \ll r$, with r being the distance between the observer and the current generating the field [2]. The dipolar approximation lends itself to describing scattering by small particles and assemblies of small particles. Importantly, the size of the assembly can very well be much larger than λ. For scattering by larger particles we refer to Reference [9].

We point out that this Chapter is not intended as a new scientific result but rather as an introduction and a reference in order to aid the unfamiliar reader in following the theoretical formalism applied throughout the remainder of this thesis. Regarding the origin of this Chapter's content we point at the quoted literature. We consider References [10, 11] and [5] together with [2] to be an ideal starting point for a practical application of the coupled-dipole model.

4.2 The dipolar approximation

Assume we have a known spatial current distribution $\boldsymbol{J}(\boldsymbol{r}', t) = \boldsymbol{J}(\boldsymbol{r}')\exp[-\mathrm{i}\omega t]$ in vacuum with a harmonic time dependence and we would like to know the electric field $\boldsymbol{E}$ generated by $\boldsymbol{J}$ at position $\boldsymbol{r}$. The vector potential $\boldsymbol{A}(\boldsymbol{r}, t) = \boldsymbol{A}(\boldsymbol{r})\exp[-\mathrm{i}\omega t]$ is then given by [1]

$$\boldsymbol{A}(\boldsymbol{r}) = \mu_0 \int_V \boldsymbol{J}(\boldsymbol{r}') \frac{\exp\left[\mathrm{i}k\left|\boldsymbol{r}-\boldsymbol{r}'\right|\right]}{4\pi\left|\boldsymbol{r}-\boldsymbol{r}'\right|} \mathrm{d}^3 r' \tag{4.1}$$

from which assuming the Lorentz gauge the electric field follows as

$$\boldsymbol{E}(\boldsymbol{r}) = \mathrm{i}\omega \left[1 + \frac{1}{k^2}\nabla\nabla\cdot\right] \boldsymbol{A}(\boldsymbol{r}), \tag{4.2}$$

where $k = n\frac{\omega}{c}$ is the wave-number in the medium.

When we consider the case where the current distribution is confined to a region much smaller than the wavelength and the observation point is far away from the source region we can immediately simplify Eq. (4.1) to read

$$\boldsymbol{A}(\boldsymbol{r}) = \mu_0 \frac{\exp[\mathrm{i}kr]}{4\pi r} \int_V \boldsymbol{J}(\boldsymbol{r}')\mathrm{d}^3 r'. \tag{4.3}$$

Performing an integration by parts and introducing the electric dipole moment

$$\boldsymbol{p} = \int_V \boldsymbol{r}' \rho(\boldsymbol{r}') \mathrm{d}^3 r' \tag{4.4}$$

the vector potential can be rewritten as

$$\boldsymbol{A}(\boldsymbol{r}) = -\mathrm{i}\omega\mu_0 \frac{\exp[\mathrm{i}kr]}{4\pi r} \boldsymbol{p}. \tag{4.5}$$

This equation constitutes the remarkable and highly useful fact that the fields generated by *any* current distribution within a volume much smaller than λ^3, observed at sufficient distance, are entirely given by the electric dipole moment of the current distribution. This observation justifies the approximation of scatterers with complex geometries by simple point dipoles as long as the scatterers are small compared to the wavelength. Importantly, the dipolar approximation describes the near fields correctly, such that the observer can go as close as desired compared to the wavelength, as long as he is still sufficiently far away measured against the spatial extent of the current distribution.

4.3 The coupled-dipole model

Restricting the discussion to dipolar interactions simplifies things tremendously. From here on, we only consider the fields generated by dipolar currents. In a homogeneous medium of refractive index n the electric field generated at position $\boldsymbol{r}_1$ by a dipole $\boldsymbol{p}$ at $\boldsymbol{r}_0$ is given by [1]

$$\boldsymbol{E}^0(\boldsymbol{p}, n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0) = \frac{\omega^2\mu_0}{4\pi r} \exp[\mathrm{i}kr] \left\{ (\hat{\boldsymbol{r}} \times \boldsymbol{p}) \times \hat{\boldsymbol{r}} + [3\hat{\boldsymbol{r}}(\hat{\boldsymbol{r}} \cdot \boldsymbol{p}) - \boldsymbol{p}] \left(\frac{1}{(kr)^2} - \frac{\mathrm{i}}{kr} \right) \right\}, \tag{4.6}$$

where $k = n\frac{\omega}{c}$ is the wavenumber in the medium of refractive index $n = \sqrt{\epsilon}$, $r = |\boldsymbol{r}_1 - \boldsymbol{r}_0|$ and $\hat{\boldsymbol{r}} = (\boldsymbol{r}_1 - \boldsymbol{r}_0)/r$.* We can generalize Eq. (4.6) to any dipolar orientation and strength $\boldsymbol{p}$ by introducing the Green function for a homogeneous medium

$$\boldsymbol{G}^0(n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0) = \left[\boldsymbol{E}(\mathbf{e}_x, n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0), \boldsymbol{E}(\mathbf{e}_y, n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0), \boldsymbol{E}(\mathbf{e}_z, n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0) \right] \tag{4.7}$$

such that

$$\boldsymbol{E}(\boldsymbol{p}, n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0) = \boldsymbol{G}(n, \omega, \boldsymbol{r}_1, \boldsymbol{r}_0) \cdot \boldsymbol{p}. \tag{4.8}$$

We have dropped the superscript in Eq. (4.8) since it can be taken as the definition of the dipolar Green function for *any* environment. Note that both dipole moments and electric fields are column vectors and the Green function is a 3×3 dyadic. It is important to keep in mind that Eq. (4.6) is the field generated by a dipole in a homogeneous medium and the field generated in another more complex environment is much more complicated. One typically separates the Green function of a complex background

*In general the refractive index $n = \sqrt{\epsilon\mu}$ depends both on the relative permittivity and permeability of the medium. Since in this thesis we are only dealing with non-magnetic materials we always assume $\mu = 1$.

system $\boldsymbol{G}_\mathrm{B} = \boldsymbol{G}^0 + \boldsymbol{G}^s$ into a sum of the free-space Green function $\boldsymbol{G}^0$ and a scattered part $\boldsymbol{G}^s$. Only for a few examples analytical solutions for $\boldsymbol{G}^s$ are known. In particular, we refer to the book by Tai [12], who calculates the Green function for a sphere, a planar interface, and an infinite cylinder of circular cross-section. Furthermore, dyadic Green functions for stratified multi-layers [2, 13], concentric spherical multi-layers [14], an eccentric spherical inclusion in a sphere [15], clusters of spheres [16] and concentric cylindrical multi-layers [17] are available but challenging to handle.

Besides the fields generated by a dipolar source, we are furthermore interested in the response of a polarizable scatterer to an incident electric field, which is in the linear approximation given by the polarizability tensor $\boldsymbol{\alpha}$, such that

$$\boldsymbol{p} = \boldsymbol{\alpha} \cdot \boldsymbol{E}. \tag{4.9}$$

Equations (4.7) and (4.9) are the ingredients necessary to set up the equations of motion for a system of N coupled dipoles. The scatterers $1,\ldots,N$ acquire dipole moments $\boldsymbol{p}_1,\ldots,\boldsymbol{p}_N$ in proportion to their polarizabilities $\boldsymbol{\alpha}_1,\ldots,\boldsymbol{\alpha}_N$ and the electric fields $\boldsymbol{E}(\boldsymbol{r}_1),\ldots,\boldsymbol{E}(\boldsymbol{r}_N)$ at their locations $\boldsymbol{r}_n$, according to the linear self-consistent set of equations [5, 10, 11]

$$\boldsymbol{p}_n = \boldsymbol{\alpha}_n \left[\boldsymbol{E}_\mathrm{in}(\boldsymbol{r}_n) + \sum_{m \neq n} \boldsymbol{G}_\mathrm{B}(\boldsymbol{r}_n, \boldsymbol{r}_m) \cdot \boldsymbol{p}_m \right], \tag{4.10}$$

where we have just rewritten Eq. (4.9) for dipole n by expressing the electric field it experiences as a superposition of the external driving field $\boldsymbol{E}_\mathrm{in}$ and the fields generated by all other polarizable particles due to the fact that they are polarized as well. By moving all dipole moments to the left of the equality sign we can now cast Eq. (4.10) in matrix form to read

$$\mathscr{P} = \boldsymbol{M}^{-1} \cdot \mathscr{E} \tag{4.11}$$

where we have concatenated the dipole moments $\boldsymbol{p}_i$ to the $3N$ component 'super-vector' for polarization $\mathscr{P}$. Equivalently $\mathscr{E}$ describes the incident field components at the particle positions, and the coupling matrix $\boldsymbol{M}$ is defined as

$$\boldsymbol{M}_{i,j} = \delta_{i,j} \boldsymbol{\alpha}_i^{-1} - (1 - \delta_{i,j}) \boldsymbol{G}_\mathrm{B}(\boldsymbol{r}_i, \boldsymbol{r}_j). \tag{4.12}$$

Note that $\boldsymbol{M}$ is of dimension $3N \times 3N$ and the sub-matrices $\boldsymbol{M}_{i,j}$ defined in Eq. (4.12) have dimension 3×3. For a scatterer with its main axes along the coordinate axes the diagonal of $\boldsymbol{M}$ holds the inverse of the polarizability tensor $\boldsymbol{\alpha}_i$ while the off-diagonal elements are given by the Green function coupling terms $\boldsymbol{G}_\mathrm{B}(\boldsymbol{r}_i, \boldsymbol{r}_j)$.

Just like Eq. (4.9) relates the dipole moment of a single scatterer to the incident electric field, Eq. (4.11) relates the polarization state of an ensemble of scatterers to the driving field, where the inverse of the coupling matrix plays the role of a polarizability tensor for the ensemble. It is instructive to take the analogy between $\boldsymbol{\alpha}$ and $\boldsymbol{M}$ even a step further by considering the procedure of diagonalization. When we consider an anisotropic particle of a conventional material we can always diagonalize its polarizability tensor. The associated coordinate transformation leads us to the

principal axes of the polarization ellipsoid, which just means that a driving field along a principal axis will never lead to a polarization of the particle in a direction orthogonal to the chosen principal axis. Equivalently, for an ensemble of particles diagonalization of the inverse coupling matrix $\boldsymbol{M}^{-1}$ leads to the 'eigen-polarizations' and 'eigen-polarizabilities' [18]. The eigen-vectors of $\boldsymbol{M}^{-1}$ can be regarded as the polarization eigen-modes of the system. We will encounter an example of a system with very peculiar polarization eigen-states in Chapter 5.

4.4 The electrodynamic polarizability

The previous section discussed the mathematical formulation and the physical meaning of the coupling matrix of a system of coupled dipoles. While we have identified the off-diagonal elements of the coupling matrix as the fields generated by the scatterers we will now search an expression for the polarizability tensor $\boldsymbol{\alpha}$ which enters the diagonal of the coupling matrix. For a truly didactic treatment of the radiation reaction discussed in the following we strongly recommend the review by Lagendijk and van Tiggelen [19].

A good starting point to find the dipole moment acquired by a spherical particle of a material described by a dielectric constant ϵ_{part} embedded in a homogeneous medium with dielectric constant ϵ_{med} when exposed to an electric field is to solve the electrostatic problem. We restrict ourself to spherical particles of an isotropic material. This approach leads to the electrostatic polarizability [9]

$$\alpha_0 = 4\pi\epsilon_0 V \frac{\epsilon_{\text{part}} - \epsilon_{\text{med}}}{\epsilon_{\text{part}} + 2\epsilon_{\text{med}}}. \tag{4.13}$$

It is now tempting to insert tabulated experimental values for $\epsilon(\omega)$ into Eq. (4.13) in order to obtain a polarizability at frequencies $\omega > 0$. Nevertheless, the obtained polarizability will remain 'electrostatic', as we will show in a moment. For certain classes of materials analytical expressions for $\epsilon(\omega)$ are known, for example a Drude metal is well described by [20]

$$\epsilon(\omega) = 1 - \frac{\omega_{\text{p}}^2}{\omega(\omega - \mathrm{i}\gamma\omega)}, \tag{4.14}$$

where ω_{p} is the plasma frequency and γ the Ohmic damping rate characterizing the material. Upon inserting Eq. (4.14) into Eq. (4.13) and for simplicity assuming the particle to be in air, we arrive at

$$\alpha_0(\omega) = 4\pi\epsilon_0 V \frac{\omega_0^2}{\omega_0^2 - \omega^2 - \mathrm{i}\gamma\omega} \tag{4.15}$$

which resembles the familiar Lorentzian line shape as the generic frequency response of any linear system with resonance frequency $\omega_0 = \omega_{\text{p}}/\sqrt{3}$. The Ohmic damping rate γ of the Drude model sets the damping of the obtained polarizability. Even though Eq. (4.15)

clearly has a frequency dependence, we still refer to it as the electrostatic polarizability, as indicated by the subscript. The reason is that having inserted a frequency dependent dielectric constant into the electrostatic polarizability Eq. (4.13) by no means ensures that the resulting expression Eq. (4.15) is physically valid. In fact, it turns out that Eq. (4.15) violates energy conservation. Accelerated charges radiate electromagnetic energy according to Larmor's formula [1] and so does any polarizable scatterer [21]. This scattering *must* show up as a loss rate in the polarizability of Eq. (4.15), which so far only contains the Ohmic damping.

We find the missing damping term by exploiting the insight that the energy loss of a scatterer equals the work done on its own current by its own electric field [2]. The apparent damping 'force' acting on the current is called Abraham-Lorentz force and has been a matter of strong debate [1, 2]. With the realization that an oscillating dipole is actually subjected to its own electric field we can rewrite Eq. (4.9) as

$$\boldsymbol{p} = \boldsymbol{\alpha}_0 \boldsymbol{E}_{\text{total}} = \boldsymbol{\alpha}_0 \left[\boldsymbol{E}_{\text{ext}} + \boldsymbol{G}_{\text{B}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\boldsymbol{p} \right], \tag{4.16}$$

where we have included the field generated by the dipole moment $\boldsymbol{p}$ at its own position via the Green function. After rearranging the terms to the form $\boldsymbol{p} = \boldsymbol{\alpha}\boldsymbol{E}_{\text{ext}}$ we find the electrodynamic polarizability

$$\boldsymbol{\alpha} = \left[\boldsymbol{\alpha}_0^{-1} - \boldsymbol{G}_{\text{B}}(\boldsymbol{r}_0, \boldsymbol{r}_0) \right]^{-1}, \tag{4.17}$$

which has an additional correction term $\boldsymbol{G}_{\text{B}}(\boldsymbol{r}_0, \boldsymbol{r}_0)$ describing the back-action of the scatterer on itself. Equation (4.17) is the scattering matrix, often referred to as t-matrix, of a single dipolar point scatterer and its expansion yields the Born series of multiple scattering [22]. Importantly, even a *single* scatterer gives rise to a *multiple* scattering series. Remarkably, with the corrected polarizability in Eq. (4.17) we have found a description of the multiple scattering problem with only a first-order scattering term. The real part $\text{Re}\,\boldsymbol{G}$ gives rise to a shift of the resonance frequency of $\boldsymbol{\alpha}$ while the imaginary part $\text{Im}\,\boldsymbol{G}$ is an additional damping term. Equation (4.17) immediately confronts us with a dramatic problem when evaluating the free-space Green-function $\boldsymbol{G}^0(\boldsymbol{r}, \boldsymbol{r})$ at the origin, since Eq. (4.6) diverges for $r = 0$. From Eqs. (4.2) and (4.5) we can read off the scalar free-space Green function [2]

$$G^0 = \frac{\omega^2 \mu_0}{4\pi r} \exp[\mathrm{i}kr], \tag{4.18}$$

which can readily be split into its real and imaginary part using Euler's formula. The real part of Eq. (4.18) indeed diverges and seems to render Eq. (4.17) useless. The problem of this divergence comes about from describing our scatterer as a true mathematical point, which we now have to approach infinitely closely. Clearly, there must be a cut-off which is sensibly chosen such that the resulting resonance frequency appears where it is experimentally found [19].

Carrying out a Taylor expansion in orders of kr before performing the spatial derivatives in order to return to the vector Green function yields for the imaginary part

of the free-space Green function

$$\mathrm{Im}\,\boldsymbol{G}^0(\boldsymbol{r}_0,\boldsymbol{r}_0) = \frac{\omega^3 n}{6\pi\epsilon_0 c^3}\mathbb{1}. \tag{4.19}$$

The imaginary part of the Green function evaluated at the origin is therefore the damping term that *has* to be included in any polarizability tensor in order to appropriately take radiation loss into account and ensure energy conservation. A complex photonic system with a scattered part of the Green function $\boldsymbol{G}^s$ changes the damping experienced by the scatterer via its imaginary part $\mathrm{Im}\,\boldsymbol{G}^s$, which enhances or reduces the free space damping $\mathrm{Im}\,\boldsymbol{G}^0$. This is the Purcell effect changing the radiative line-width of a *scatterer*. The real part $\mathrm{Re}\,\boldsymbol{G}^s$ shifts the resonance frequency of the scatterer. Thanks to the tensorial nature of $\boldsymbol{G}$ the polarizability of an isotropic scatterer can acquire an anisotropy due to its environment entering in the correction according to Eq. (4.17).

A straightforward recipe to include radiation damping and obtain a bona-fide electrodynamic polarizability $\boldsymbol{\alpha}$ in a homogeneous medium of refractive index n for any chosen electrostatic $\boldsymbol{\alpha}_0$ is therefore [11, 23]

$$\boldsymbol{\alpha}^{-1} = \boldsymbol{\alpha}_0^{-1} - \mathrm{i}\frac{1}{6\pi\epsilon_0}\frac{\omega^3}{c^3} n\,\mathbb{1}. \tag{4.20}$$

For spheres, the correction in Eq. (4.20) is sometimes amended by a further depolarization factor [24], which leads to a line-shift to the red with increasing particle size but is not strictly necessary to conserve energy.

4.5 Observables and the optical theorem

With the coupled-dipole model we have outlined a consistent electrodynamic framework to describe scattering. In the present section we derive observables that are experimentally accessible. By purely energetic considerations we will arrive at expressions for the extinction and scattering cross-sections of a single dipolar scatterer in a homogeneous medium as well as an expression for the optical theorem for a single dipolar scatterer in any environment.

We consider a single dipolar current source $\boldsymbol{j} = \dot{\boldsymbol{p}} = \boldsymbol{j}_0 \exp[-\mathrm{i}\omega t]\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)$ located at $\boldsymbol{r}_0$. For the moment we do not worry about how this current is generated. We apply the time-averaged form of Poynting's theorem to consider the flux of energy through an arbitrary surface ∂V enclosing only our dipolar current and no lossy material. Poynting's theorem relates the time-averaged flux of electromagnetic energy described by the Poynting vector $\langle \boldsymbol{S}\rangle = \frac{1}{2}\mathrm{Re}\left\{\boldsymbol{E}\times\boldsymbol{H}^*\right\}$ through the chosen surface to the fields and currents within the enclosed volume via [2]

$$\int_{\partial V}\langle \boldsymbol{S}\rangle\,\mathrm{d}\boldsymbol{A} = -\frac{1}{2}\int_V \mathrm{Re}\left\{\boldsymbol{j}^\dagger\boldsymbol{E}\right\}\mathrm{d}V, \tag{4.21}$$

where $(\cdot)^\dagger$ denotes the Hermitian conjugate and we imply usual matrix multiplication. Expressing the fields generated by our dipole $\boldsymbol{p}$ via the Green function of the embedding system we arrive at the power emitted by our source into the (possibly lossy)

environment

$$P_{\mathrm{em}} = \frac{1}{2}\omega \boldsymbol{p}^{\dagger} \mathrm{Im}\left[\boldsymbol{G}(\boldsymbol{r}_0, \boldsymbol{r}_0)\right] \boldsymbol{p}. \tag{4.22}$$

In the special case of a homogeneous lossless medium of refractive index n, Eq. (4.22) turns into the familiar form of Larmor's formula $P = \frac{\omega^4 n}{12\pi\epsilon_0 c^3} |\boldsymbol{p}|^2$ for the radiated power of a dipole in a homogeneous medium upon inserting $\mathrm{Im}\,\boldsymbol{G}^0$ from Eq. (4.19). Note that in the case of an environment including lossy constituents Eq. (4.22) describes the sum of the power radiated into the continuum and the power absorbed by the environment, provided you can draw a closed surface around the source *without* enclosing any lossy material.

So far, we have not specified what creates the dipole moment $\boldsymbol{p}$. While Eq. (4.22) holds for *any* environment, in order to derive the scattering and absorption cross-section of a single particle we now consider the case of a single isotropic scatterer with polarizability α in a homogeneous lossless medium driven by a plane wave $\boldsymbol{E}_{\mathrm{in}}$. We insert $\boldsymbol{p} = \alpha \boldsymbol{E}$ into Eq. (4.22) to calculate the power scattered by the particle and remember that the time-averaged Poynting vector along the propagation direction $\langle S \rangle = \frac{1}{2}\sqrt{\frac{\epsilon_0}{\mu_0}} n |\boldsymbol{E}|^2$ gives the incoming power. By dividing the scattered power by the incoming power flux density we obtain the scattering cross-section

$$\sigma_{\mathrm{scat}} = \frac{\omega^4}{6\pi\epsilon_0^2 c^4} |\alpha|^2 . \tag{4.23}$$

For a weak scatterer, i.e. far away from any resonance in α, the wavelength dependence shows the famous λ^{-4} behavior of Rayleigh scattering, which is one main reason why the sky appears blue. This dependence can be obtained from a simple dimensional analysis [25]. Importantly, we have arrived at the scattered power by considering the work that is done on the dipole moment by its *own* field. In contrast, the energy removed by the scatterer from the incoming beam must equal the work that is done on the dipole moment by the *incoming* field. The cycle averaged extinction is therefore [10]

$$P_{\mathrm{ext}} = \langle \mathrm{Re}\, \boldsymbol{E}_{\mathrm{in}} \cdot \mathrm{Re}\, \boldsymbol{j} \rangle = \frac{1}{2}\omega \mathrm{Im}\,(\boldsymbol{E}_{\mathrm{in}}^{\dagger} \boldsymbol{p}), \tag{4.24}$$

which leads to the extinction cross-section of a dipolar scatterer in a homogeneous medium

$$\sigma_{\mathrm{ext}} = \frac{\omega}{\epsilon_0 c n} \mathrm{Im}\,\alpha. \tag{4.25}$$

We note after comparing Eqs. (4.23) and (4.25) that since $\alpha \propto V$ the ratio of scattered to extinct power, commonly referred to as the albedo [19], vanishes for small volumes, such that small particles practically only absorb.

Importantly, in any environment described by $\boldsymbol{G}$, energy conservation requires that the scattered power Eq. (4.22) can never exceed the extinct power Eq. (4.24), such that

$$\boldsymbol{\alpha}^{\dagger} \mathrm{Im}\, \boldsymbol{G} \boldsymbol{\alpha} \leq \frac{1}{2\mathrm{i}} \left(\boldsymbol{\alpha} - \boldsymbol{\alpha}^{\dagger} \right). \tag{4.26}$$

The relation Eq. (4.26) is a general form of the optical theorem that restricts the polarizability $\boldsymbol{\alpha}$ to conserve energy [26]. In the special case of a lossless and homogeneous medium with refractive index n and an isotropic scatterer with polarizability α Eq. (4.26) reduces to [27]

$$\frac{n\omega^3}{6\pi\epsilon_0 c^3}|\alpha|^2 \leq \operatorname{Im}\alpha. \tag{4.27}$$

The correction of the polarizability given in Eq. (4.20) makes sure that the optical theorem Eq. (4.27) is obeyed and energy is conserved. The equality sign in Eqs. (4.26) and (4.27) holds for the case of any hypothetical scatterer without material loss, i.e. $\gamma = 0$, on resonance. More importantly for practical cases, the radiation damping term entering the denominator of the expression for the polarizability according to Eq. (4.17) scales with the particle volume, such that for large scatterers the radiation damping will exceed the material damping. The line-width of the polarizability of plasmonic particles larger than about 40 nm in diameter is typically dominated by radiation losses [28].

Importantly, the electromagnetic environment entering Eq. (4.26) via the Green function $\boldsymbol{G}$ bounds the scattering strength of any dipole to the *unitary limit*. Let us consider an isotropic scatterer in vacuum, where we can combine Eqs. (4.25) and (4.27) to obtain the maximally possible extinction cross-section of *any* dipolar scatterer $\sigma_{\text{ext}}^{\text{UL}} = \frac{3}{2\pi}\lambda^2$. We will reencounter the unitary limit in the context of assemblies of scatterers in Chapter 5 and its repercussions in a complex environment will be of paramount importance in Chapter 6.

We have just reminded ourself how to calculate the extinction and scattering cross-sections of single particles. The value and success of the coupled-dipole model relies on the fact that it allows to calculate extinction, scattering, and absorption cross-sections of clusters of particles, as well as eigen-modes and radiation patterns, also for clusters of particles [5, 10, 11, 18, 29]. As we will illustrate in the next section, also the LDOS is easily available in a coupled-dipole approach. The coupled-dipole formalism has recently been extended to magneto-electric scatterers [27]. The typical procedure to obtain any observable is to first specify the particle positions and their electrodynamic polarizabilities. At this point the coupling matrix $\boldsymbol{M}$, given in Eq. (4.12), is fully defined and has to be inverted. To find the dipole moments for any driving field remains a simple matrix multiplication according to Eq. (4.11). With the knowledge of the dipole moments resulting from a specific driving field all desired observables can be derived. For example, the extinction cross-section of a particle cluster is obtained from choosing a plane wave as a driving field and summing the power dissipated by that driving field by acting on the resulting individual dipole moments according to Eq. (4.24). To obtain the scattering cross-section of a particle cluster we calculate the Poynting vector of the fields generated by the individual dipole moments through a hypothetical sphere. This procedure can be carried out with limited computational effort by choosing the sphere sufficiently large and applying a far-field approximation for the dipole radiation. With the same recipe we can naturally also calculate differential scattering cross-sections and thereby radiation patterns of particle clusters.

Regarding the driving field, we are of course not limited to plane waves. Most interesting for the purpose of this thesis is certainly to choose a dipolar point source to

drive an ensemble of scatterers to model the field emitted by a dipolar source in the vicinity of a cluster of scatterers. The dipole field generated by that point source, given in Eq. (4.6), then enters Eq. (4.11) as the driving field and the total radiated field is the superposition of the driving field and the fields generated by the induced dipole moments of the scatterers calculated according to Eq. (4.11).

4.6 Spontaneous-emission rate enhancement

Most interesting for the purpose of this thesis is to calculate the LDOS at a particular position with respect to an ensemble of point scatterers. Xu, Lee, and Yariv [30] have shown that the spontaneous-emission rate of a quantum-mechanical two-level system with transition frequency ω is proportional to the power dissipated by a constant-current source of the same frequency [2]. We can therefore calculate the spontaneous-emission rate *enhancement* via the enhancement in power dissipated by a classical constant-current source in the complex photonic environment as compared to a reference system. To this end, we revert to the expression for the power dissipated by an oscillating dipole in Eq. (4.24), under the assumption that $\mathrm{Im}\,\boldsymbol{G}$ does not significantly change across the natural line-width of the quantum emitter [30, 31]. Upon splitting the Green function into its free and scattered parts the decay-rate enhancement with respect to vacuum is given by

$$A = 1 + \frac{\boldsymbol{p}^\dagger \mathrm{Im}\,\boldsymbol{G}^{\mathrm{s}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\boldsymbol{p}}{\boldsymbol{p}^\dagger \mathrm{Im}\,\boldsymbol{G}^{\mathrm{vac}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\boldsymbol{p}}. \tag{4.28}$$

From Eq. (4.28) we can immediately appreciate that the rate enhancement (or suppression) of a spontaneous emitter in a complex photonic system is a result of the radiation reaction of the emitter's own field scattered by the environment and returning with a phase shift to perform work on the source, thereby increasing (or decreasing) the resistance of the vacuum. This picture merits the interpretation of the LDOS as an impedance experienced by a quantum emitter [32, 33]. Importantly, Eq. (4.28) allows to calculate the decay-rate enhancement, and therefore the LDOS, in any photonic environment whose Green function is known, despite that system possibly exhibiting material losses, which renders literal counting of the states impossible [34, 35]. The strength of Eq. (4.28) is that it allows to determine the LDOS by evaluating $\mathrm{Im}\,\boldsymbol{G}$ only at a single point, namely the origin. It however hides the different contributions to the LDOS since the dissipated power calculated according to Eq. (4.28) contains all contributions to the LDOS, both radiative and non-radiative, due to the environment. If we desire to separate the rate enhancement due to radiative decay enhancement we will have to integrate the radiated power in the far field.

Finally, we would like to point out that the community of spontaneous-emission control, including ourself, handles the term LDOS rather sloppily. LDOS is used to refer to the energy and volume density of states, but also to the imaginary part of the Green function $\mathrm{Im}\,\boldsymbol{G}(\boldsymbol{r}, \boldsymbol{r})$, its component projected on a certain dipole orientation $\hat{\boldsymbol{p}}^\dagger \mathrm{Im}\,\boldsymbol{G}^{\mathrm{vac}}(\boldsymbol{r}_0, \boldsymbol{r}_0)\hat{\boldsymbol{p}}$, its trace $\mathrm{tr}\{\mathrm{Im}\,\boldsymbol{G}\}$ or any of these quantities normalized to the corresponding value in vacuum. Typically, no confusion arises from the context.

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