Spontaneous emission dynamics - Lecture notes

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Abstract

These notes summarize the content of lectures that have been given in different Master courses and summer schools. The purpose is to describe the emission dynamics of a dipole emitter in a structured environment. The focus is on the weak-coupling regime, in which the main feature is the dependence of the spontaneous decay rate on the environment, known as the Purcell effect. The local density of states is introduced as a central concept in the description.

1 Introduction

These notes introduce the theoretical concepts needed to describe the spontaneous emission dynamics of an emitter embedded in a structured environment (surface, cavity, nanoantenna...) in the weak coupling regime. A feature of this regime is the irreversible transition of the emitter from an excited state to the ground state by photon emission (a process also known as fluorescence).

The spontaneous emission dynamics is characterized by the lifetime τ of the excited state, or equivalently the decay rate $\Gamma = 1/\tau$. The decay rate is directly observable, as shown in Fig. 1.

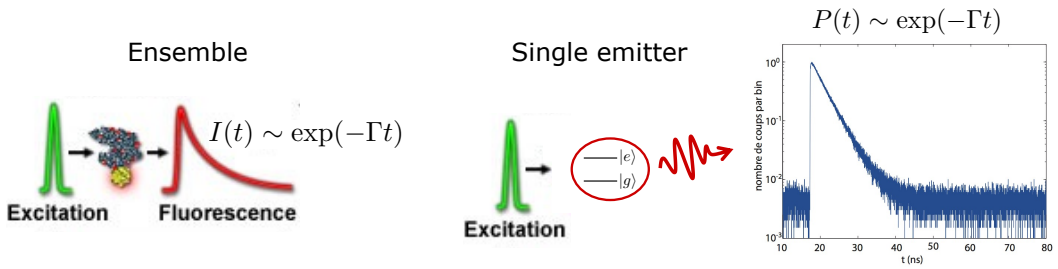

Figure 1: Measurement of the fluorescence decay rate Γ. On an ensemble (for example fluorescent molecules in solution), the molecules are initially excited by a short laser pulse and the fluorescence intensity $I(t)$ decays over time as $\exp(-\Gamma t)$. For a single emitter, one needs to repeat many cycles of excitation by a short laser pulse and detection of the emitted photon, and build a histogram of the delays between excitation and photon detection, as shown in the right panel. The probability $P(t)$ for the emitter to remain excitated at time t decays as $\exp(-\Gamma t)$.

It is known that the spontaneous decay rate is not an intrinsic property of an emitter, but depends on the environment. An example of an early measurement is shown in Fig. 2. The lifetime of the emitters is seen to depend substantially on the distance to a silver mirror [1].

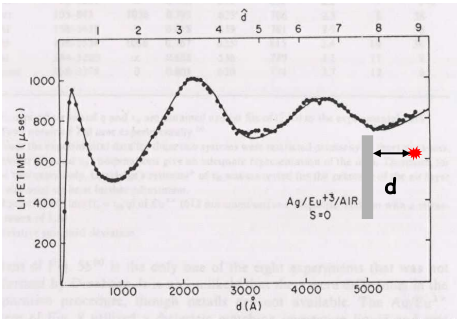

Figure 2: Fluorescence lifetime of emitters (here europium ions) in front of a silver mirror versus the distance d. One observes oscillations resulting from the interaction with the reflected field, and a strong decrease at short distance due to non radiative coupling (absorption). The solid line is a theoretical model using concepts similar to those introduced in the present note. Adapted from [1].

Here we will introduce the basic concepts and methods needed to describe the dependence of the decay rate on the local environment of the emitter. We will favor an approach connecting the decay rate to the power emitted by a classical dipole, as described for example in [2]. The interested reader could find an alternative approach based on a scattering formalism in Ref. [3], that allows one to recover the results included in the present notes but from a different perspectives, and that also includes other regimes (strong coupling, energy transfer) not discussed here.

2 Power radiated by classical dipole source

2.1 General expression for a monochromatic source

Consider a monochromatic source radiating in an arbitrary environment at frequency ω (we assume that all fields and sources have a time dependence of the form $\exp(-i\omega t)$). The source is described by a current density with complex amplitude $\mathbf{j}$ occupying a volume V. The time-averaged power transferred from the source to the environment is

$$P = -\frac{1}{2}\,\mathrm{Re}\int \mathbf{j}^*(\mathbf{r})\cdot\mathbf{E}(\mathbf{r})\,d^3r\,, \tag{1}$$

where the superscript $*$ denotes the complex conjugate, and the integral is extended to the volume V of the source. For an electric dipole source located at the point $\mathbf{r}_s$, the current density is $\mathbf{j}(\mathbf{r}) = -i\omega\,\mathbf{p}\,\delta(\mathbf{r}-\mathbf{r}_s)$, with $\mathbf{p}$ the dipole moment. The emitted power takes the simple form

$$P = \frac{\omega}{2}\,\mathrm{Im}\left[\mathbf{p}^*\cdot\mathbf{E}(\mathbf{r}_s)\right]\,. \tag{2}$$

2.2 The response of the environment: Green's function

In an arbitrary environment described by a dielectric function $\varepsilon(\mathbf{r})$ (we assume that the media surrounding the source are non-magnetic), the electric field obeys the vector form of Helmholtz's equation:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \varepsilon(\mathbf{r}) k_0^2 \, \mathbf{E}(\mathbf{r}) = i\mu_0 \omega \, \mathbf{j}(\mathbf{r}) \,, \tag{3}$$

where $k_0 = \omega/c$ with c the speed of light in vacuum. By making use of the concept of Green's function, the general solution of the above equation can be written as an integral connecting the electric field to the source $\mathbf{j}$. Formally, the (tensor) Green's function is the solution to

$$\nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) - \varepsilon(\mathbf{r}) k_0^2 \, \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{I} \, \delta(\mathbf{r} - \mathbf{r}') \,, \tag{4}$$

where $\mathbf{I}$ is the unit second-rank tensor. Physically, the Green's function describes the radiation at the point $\mathbf{r}$ generated by a point source located at $\mathbf{r}'$. One usually chooses the solution satisfying an outgoing wave condition when $|\mathbf{r} - \mathbf{r}'| \to \infty$, meaning that far from the source point the radiated field behaves as an outgoing spherical wave of the form $\exp(ik_0|\mathbf{r} - \mathbf{r}'|)/(|\mathbf{r} - \mathbf{r}'|)$. For a deeper introduction to Green's functions, see for example the dedicated chapters in [4, 5].

Making use of the Green's function, the solution to Eq. (3) can be written

$$\mathbf{E}(\mathbf{r}) = i\mu_0 \omega \int \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \, \mathbf{j}(\mathbf{r}') \, d^3 r' \,, \tag{5}$$

which can be understood as a linear superposition of the fields emitted by each point of the source. Note that here $\mathbf{G}$ is a second-rank tensor with components G_{jk} such that the vector components of $\mathbf{E}$ and $\mathbf{j}$ are related by

$$E_j(\mathbf{r}) = i\mu_0 \omega \int \sum_k G_{jk}(\mathbf{r}, \mathbf{r}', \omega) \, j_k(\mathbf{r}') \, d^3 r' \,. \tag{6}$$

In the case of the emission by an electric dipole source located at the point $\mathbf{r}_s$, the radiated field takes the form

$$\mathbf{E}(\mathbf{r}) = \mu_0 \omega^2 \mathbf{G}(\mathbf{r}, \mathbf{r}_s, \omega) \, \mathbf{p} \,, \tag{7}$$

and Eq. (2) can be rewritten as

$$P = \frac{\mu_0 \omega^3}{2} \, |\mathbf{p}|^2 \, \mathrm{Im} \left[\mathbf{u} \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega) \mathbf{u} \right] \,. \tag{8}$$

In this equation $\mathbf{u} = \mathbf{p}/|\mathbf{p}|$ is the unit vector defining the orientation of the emitting dipole. One advantage of this representation is that the source power ($|\mathbf{p}|^2$) and the electromagnetic response of the environment ($\mathbf{G}$) factorize. We also note that the emitted power is driven by the imaginary part of the Green's function.

2.3 Particular case: Emission in free space

In free space, the Green's function $\mathbf{G}_0$ is the solution to Eq. (4) with $\varepsilon(\mathbf{r}) = 1$ everywhere. The solution satisfying the outgoing wave condition is [4, 5]

$$\mathbf{G}_0(\mathbf{r}, \mathbf{r}', \omega) = \frac{\exp(ik_0 R)}{4\pi R} \left[\mathbf{I} - \hat{\mathbf{R}} \otimes \hat{\mathbf{R}} - \left(\frac{1}{ik_0 R} + \frac{1}{k_0^2 R^2} \right) \left(\mathbf{I} - 3\hat{\mathbf{R}} \otimes \hat{\mathbf{R}} \right) \right] - \frac{\mathbf{I}}{3k_0^2} \delta(\mathbf{R}) \,, \tag{9}$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}'$, $R = |\mathbf{R}|$ and $\hat{\mathbf{R}} = \mathbf{R}/R$ is the unit vector along $\mathbf{R}$. We use the notation $\otimes$ for the dyadic product.[1]

The calculation of the imaginary part in the limit $\mathbf{r} = \mathbf{r}'$ gives

$$\mathrm{Im}\,\mathbf{G}_0(\mathbf{r}, \mathbf{r}, \omega) = \frac{k_0}{6\pi}\mathbf{I}, \tag{10}$$

the derivation of this result being left as an exercise. Inserting the above expression into Eq. (8) leads to

$$P_0 = \frac{\omega^4}{12\pi\epsilon_0 c^3}\,|\mathbf{p}|^2\,, \tag{11}$$

which is the well-known expression of the power radiated by en electric dipole in vacuum.

3 Spontaneous decay rate of a quantum emitter

3.1 Decay rate from quantum perturbation theory

Consider a two-level atom with excited and ground states $|e\rangle$ and $|g\rangle$, respectively. We denote by ω the transition frequency such that $\omega = (E_e - E_g)/\hbar$, with E_e and E_g the energies of the two states. The spontaneous decay rate Γ of an atom initially in the excited state can be obtained from perturbation theory. For an atom located at the point $\mathbf{r}_s$, the result takes the form

$$\Gamma = \frac{2\mu_0\omega^2}{\hbar}\,|\mathbf{p}_{eg}|^2\,\mathrm{Im}\left[\mathbf{u}\cdot\mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}\right]\ . \tag{12}$$

Here $\mathbf{p}_{eg}$ is the transition dipole, defined as the matrix element $\mathbf{p}_{eg} = \langle g|\mathbf{D}|e\rangle$ of the transition dipole operator $\mathbf{D}$, and $\mathbf{u} = \mathbf{p}_{eg}/|\mathbf{p}_{eg}|$ defines the orientation of the dipole, as in the classical case. Deriving the above equation in a quantum framework is beyond the scope of these lecture notes (see for example [6, 7] for a detailed treatment). Instead, we will show that Eq. (12) can be deduced from the classical emitted power in a correspondence fashion.

3.2 A classical to quantum correspondence

We propose here to deduce the expression of the quantum decay rate Γ from the expression (8) of the power radiated by a classical electric dipole. We follow an approach introduced by Born in Ref. [8], based on correspondence arguments.

First, we change in Eq. (8) the classical dipole moment $\mathbf{p}$ into the quantum transition dipole $\mathbf{p}_{eg}$, and the classical frequency ω into the Bohr frequency $\omega = (E_e - E_g)/\hbar$, to obtain the power emitted per unit time in the quantum picture. In the replacement of the transition dipole, we need to account for a factor of 2, due to the fact that only positive frequencies have to be used (in quantum mechanics, a clear meaning is given to either positive or negative frequencies).[2] Second, we write that for an atom initially in the excited state, the emitted power is $\Gamma\hbar\omega$.

[1] For any two vectors $\mathbf{u}$ and $\mathbf{v}$ the components of the tensor $(\mathbf{u}\otimes\mathbf{v})$ are $(\mathbf{u}\otimes\mathbf{v})_{ij} = u_i v_j$. Applying this tensor to a vector $\mathbf{w}$ leads to $(\mathbf{u}\otimes\mathbf{v})\mathbf{w} = (\mathbf{v}\cdot\mathbf{w})\mathbf{u}$.

[2] The frequency spectrum of the electric dipole $\mathbf{p}(t)$ is expressed by the Fourier transform $\mathbf{p}(t) = \int_{-\infty}^{+\infty}\mathbf{p}(\omega)\exp(-i\omega t)d\omega$. Since $\mathbf{p}(t)$ is real, $\mathbf{p}(-\omega) = \mathbf{p}^*(\omega)$ and $\mathbf{p}(t) = 2\mathrm{Re}\int_0^{+\infty}\mathbf{p}(\omega)\exp(-i\omega t)d\omega$. We see that the spectrum reduced to positive frequencies contains a factor of 2.

These two steps lead to the conclusion that $\Gamma = P/\hbar\omega$, where P is given by Eq. (8) with the two replacements above, and we find that

$$\Gamma = \frac{2\mu_0\omega^2}{\hbar} |\mathbf{p}_{eg}|^2 \, \mathrm{Im}\left[\mathbf{u} \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}\right] , \tag{13}$$

which agrees with Eq. (12).

3.3 Decay rate in free space

The decay rate in free space Γ_0 is obtained by using the vacuum Green's function $\mathbf{G}_0$ in Eq. (13), and making use of Eq. (10). We find that

$$\Gamma_0 = \frac{\omega^3}{3\pi\hbar\epsilon_0 c^3} |\mathbf{p}_{eg}|^2 , \tag{14}$$

which is the known result for the decay rate of a two-level atom in vacuum.

3.4 Normalized decay rate

The normalized decay rate Γ/Γ_0 is readily obtained from the two equations above:

$$\frac{\Gamma}{\Gamma_0} = \frac{6\pi}{k_0} \mathrm{Im}\left[\mathbf{u} \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}\right] . \tag{15}$$

The right-hand side describes the change in the decay rate of a quantum emitter due to the environment.

It can be easily verified that for the radiation from a classical dipole source, normalizing the emitted power P by the free space power P_0 leads to the same result, namely

$$\frac{P}{P_0} = \frac{6\pi}{k_0} \mathrm{Im}\left[\mathbf{u} \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}\right] . \tag{16}$$

In other words, the change in the emitted power P of a classical dipole with fixed dipole moment $\mathbf{p}$ due to the environment is the same as the change in the decay rate of a quantum emitter emitting at the same frequency and located at the same position. We conclude that changes in decay rates can be computed from purely classical concepts.

4 Local density of states

We have seen that the internal dynamics of the emitter and the influence of the environment can be factorized in the expression of the decay rate, the role of the environment being described by the imaginary part of the Green's function (more precisely by its projection on the orientation $\mathbf{u}$ of the emitting dipole). The role of the environment is often described in terms of the local density of states (LDOS) that we will introduce in this section. For a more detailed presentation see Refs. [2, 4, 5].

4.1 LDOS and Green's function

We start by defining the LDOS of the electromagnetic field as

$$\rho(\mathbf{r},\omega) = \frac{2\omega}{\pi c^2}\,\mathrm{Im}\left[\mathrm{Tr}\,\mathbf{G}(\mathbf{r},\mathbf{r},\omega)\right]\,, \tag{17}$$

with Tr denoting the trace of a tensor. More precisely, this expression defines the electric contribution to the LDOS, relevant to describe the decay rate of emitters with an electric dipole transition. For magnetic dipole transitions, a magnetic LDOS can be introduced [2].

We also define a projected LDOS, relevant to describe the decay rate of emitters with a given orientation $\mathbf{u}$ of the transition dipole as follows:

$$\rho_{\mathbf{u}}(\mathbf{r},\omega) = \frac{2\omega}{\pi c^2}\,\mathrm{Im}\left[\mathbf{u}\cdot\mathbf{G}(\mathbf{r},\mathbf{r},\omega)\mathbf{u}\right]\,. \tag{18}$$

The full LDOS $\rho = \rho_{\mathbf{u}_x} + \rho_{\mathbf{u}_y} + \rho_{\mathbf{u}_z}$ sums up the three orientational degrees of freedom.

In terms of the projected LDOS, the spontaneous decay rate of a quantum emitter located at the point $\mathbf{r}_s$, emitting at a frequency ω, and with a transition dipole oriented along the unit vector $\mathbf{u}$, can be written as

$$\Gamma = \frac{\pi\omega}{\hbar\epsilon_0}\,|\mathbf{p}_{eg}|^2\,\rho_{\mathbf{u}}(\mathbf{r}_s,\omega)\,. \tag{19}$$

This expression takes the form of Fermi's golden rule (the larger the density of states, the larger the decay rate).

4.2 Physical picture: A local counting of electromagnetic modes

Defining the LDOS from the imaginary part of the Green's function is very general, but the fact that the LDOS defined this way performs a counting of the electromagnetic modes is somehow hidden. To make it explicit, we consider a non-absorbing and non-dispersive medium described by a real-valued and frequency-independent dielectric function $\varepsilon(\mathbf{r})$ enclosed in a cavity (no radiation losses). In this particular case, we can introduce a set of eigenmodes for the electromagnetic field (we follow the approach in Ref. [9]).

We consider the eigenmodes $\mathbf{e}_n$ with eigenfrequencies ω_n which obey the vector Helmholtz equation

$$\nabla\times\nabla\times\mathbf{e}_n(\mathbf{r}) - \varepsilon(\mathbf{r})\frac{\omega_n^2}{c^2}\,\mathbf{e}_n(\mathbf{r}) = 0\,. \tag{20}$$

This equation can be rewritten as

$$\left[\frac{1}{\sqrt{\varepsilon(\mathbf{r})}}\nabla\times\nabla\times\frac{1}{\sqrt{\varepsilon(\mathbf{r})}}\right]\mathbf{u}_n(\mathbf{r}) - \frac{\omega_n^2}{c^2}\,\mathbf{u}_n(\mathbf{r}) = 0 \tag{21}$$

with $\mathbf{u}_n(\mathbf{r}) = \sqrt{\varepsilon(\mathbf{r})}\mathbf{e}_n(\mathbf{r})$. Equation (21) defines an eigenvalue problem for a Hermitian operator, that admits real eigenfrequencies ω_n and eigenfunctions satisfying the orthonormality condition

$$\int \mathbf{u}_m(\mathbf{r})\cdot\mathbf{u}_n^*(\mathbf{r})\,d^3r = \delta_{mn}\,. \tag{22}$$

The orthogonality condition for the eigenmodes solution to Eq. (20) takes the form

$$\int \varepsilon(\mathbf{r})\,\mathbf{e}_m(\mathbf{r})\cdot\mathbf{e}_n^*(\mathbf{r})\,d^3r = \delta_{mn}\ . \tag{23}$$

Our goal is to expand the Green's function satisfying Eq. (4) on the basis of the eigenmodes $\mathbf{e}_n$ in the form

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \sum_n \mathbf{e}_n(\mathbf{r})\otimes\mathbf{g}_n(\mathbf{r}')\,, \tag{24}$$

where the coefficients $\mathbf{g}_n$ need to be determined.

Inserting (24) into (4) yields

$$\sum_n \left[\nabla\times\nabla\times\mathbf{e}_n(\mathbf{r}) - \varepsilon(\mathbf{r})\frac{\omega^2}{c^2}\,\mathbf{e}_n(\mathbf{r})\right]\otimes\mathbf{g}_n(\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')\mathbf{I} \tag{25}$$

which, using Eq. (20), can be transformed into

$$\sum_n \left(\frac{\omega_n^2}{c^2} - \frac{\omega^2}{c^2}\right)\varepsilon(\mathbf{r})\,\mathbf{e}_n(\mathbf{r})\otimes\mathbf{g}_n(\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')\mathbf{I}\ . \tag{26}$$

Multiplying both sides by $\mathbf{e}_m^*(\mathbf{r})$, integrating over $\mathbf{r}$ and using the orthogonality condition (23) leads to

$$(\omega_n^2-\omega^2)\,\mathbf{g}_n(\mathbf{r}') = c^2\mathbf{e}_n^*(\mathbf{r}')\,. \tag{27}$$

The general solution to the above equation is of the form

$$\mathbf{g}_n(\mathbf{r}') = c^2\mathbf{e}_n^*(\mathbf{r}')\left[\mathrm{P}\left(\frac{1}{\omega_n^2-\omega^2}\right) + A\delta(\omega-\omega_n) + B\delta(\omega+\omega_n)\right]\,, \tag{28}$$

where A and B are constants and P denotes the principal value. To determine the constants, we note that

$$\frac{1}{\omega_n^2-\omega^2} = \frac{1}{2\omega_n}\left(\frac{1}{\omega_n-\omega}+\frac{1}{\omega_n+\omega}\right) \tag{29}$$

and make use of the identity

$$\lim_{\eta\to 0}\frac{1}{x-x_0-i\eta} = \mathrm{PV}\frac{1}{x-x_0} + i\pi\,\delta(x-x_0)\,. \tag{30}$$

This leads to $A = i\pi/(2\omega)$ and $B = -i\pi/(2\omega)$. Finally, from Eqs. (24) and (28), we obtain the eigenmodes expansion of the Green's function:

$$\mathbf{G}(\mathbf{r},\mathbf{r}',\omega) = \sum_n c^2\,\mathbf{e}_n(\mathbf{r})\otimes\mathbf{e}_n^*(\mathbf{r}')\left[\mathrm{P}\left(\frac{1}{\omega_n^2-\omega^2}\right) + \frac{i\pi}{2\omega_n}\delta(\omega-\omega_n)\right]\ . \tag{31}$$

We have dropped the term proportionnal to $\delta(\omega+\omega_n)$ since we will define the LDOS for positive frequencies only, and this term does not contribute.[3]

[3]The Green's function has Hermitian symmetry $\mathbf{G}(\mathbf{r},\mathbf{r}',-\omega) = \mathbf{G}^*(\mathbf{r},\mathbf{r}',\omega)$ in the frequency domain and the spectrum can be restricted to positive frequencies.

From this expression, we readily deduce that

$$\mathrm{Im}\left[\mathrm{Tr}\,\mathbf{G}(\mathbf{r},\mathbf{r},\omega)\right] = \frac{\pi c^2}{2\omega}\sum_n |\mathbf{e}_n(\mathbf{r})|^2 \delta(\omega-\omega_n)\,, \tag{32}$$

where ω_n had been replaced by ω in the prefactor, keeping the result unchanged due to the presence of the delta function. We immediately find that the LDOS defined in Eq. (17) reads

$$\rho(\mathbf{r},\omega) = \sum_n |\mathbf{e}_n(\mathbf{r})|^2\,\delta(\omega-\omega_n)\ . \tag{33}$$

This expression gives insight into the meaning of the LDOS. We see that $\rho(\mathbf{r},\omega)$ counts the number of eigenfrequencies in an infinitely small frequency interval around ω (the LDOS is a spectral density), weighted by the contribution of each eigenmode at the point $\mathbf{r}$. Large (small) LDOS means many (few) modes and/or modes with large (weak) intensities at a given points, as illustrated in Fig. 3. It is important to keep in mind that an explicit relation of the type (33) makes sense when a set of eigenmodes can be properly defined, which occurs only in a few particular cases such as a non-dissipative medium enclosed in a cavity. In the most general situation of a dissipative system (with material absorption and/or radiation losses), no such simple expansion exist. Nevertheless, we can still define a LDOS based on expression (17) or (18) and use it to describe spontaneous emission.

Figure 3: Illustrating the concept of LDOS, with roads representing eigenmodes.

4.3 LDOS and the impedance of an antenna

Connecting the change in the decay rate Γ/Γ_0 to a change in the LDOS is the common approach in quantum theory. The equivalence with the change P/P_0 in the power emitted by a classical dipole emitter (or antenna) calls for another point of view. Indeed, in antenna theory, the change in the emitted power is usually associated to a change in the impedance. Here we show that LDOS and impedance are actually related by a simple expression. A deeper discussion of this relation, and its implications, can be found in Ref. [10].

Consider a wire antenna, as depicted in the left panel in Fig. 4. The antenna is fed by a monochromatic current I (frequency ω) produced by an ideal current source, and we denote by U the voltage between the two extremities of the antenna. We assume an ideal antenna with radiation losses only.

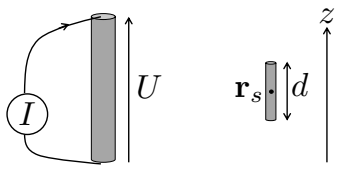

Figure 4:

The time-averaged power emitted by the antenna can be written as

$$P = \frac{1}{2}\mathrm{Re}(UI^*) = \frac{1}{2}\mathrm{Re}(Z)\,|I|^2\,, \tag{34}$$

where $Z = U/I$ is the radiation impedance of the antenna.

For an electric dipole antenna, taken to be an infinitely thin wire with length $d \ll \lambda$, located at the point $\mathbf{r}_s$, and oriented along the z-direction (as depicted in the right panel in Fig. 4), the emitted power can be expressed using Eq. (8). We find that

$$P = \frac{\mu_0\omega^3}{2}|\mathbf{p}|^2\mathrm{Im}\left[\mathbf{u}_z \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}_z\right]\,, \tag{35}$$

where $\mathbf{u}_z$ is the unit vector along the z-axis, $\mathbf{p} = p\mathbf{u}_z$ is the dipole moment of the antenna, and $\mathbf{G}$ is the Green's function describing the electrodynamic response of the environment.

Using the fact that $-i\omega\mathbf{p} = \int \mathbf{j}(\mathbf{r})d^3r = Id\,\mathbf{u}_z$, we find that the dipole moment and the current in the antenna are related by $|\mathbf{p}|^2 = |I|^2d^2/\omega^2$. Making use of Eqs. (34) and (35), we obtain

$$\mathrm{Re}(Z) = \mu_0\omega\, d^2\,\mathrm{Im}\left[\mathbf{u}_z \cdot \mathbf{G}(\mathbf{r}_s, \mathbf{r}_s, \omega)\mathbf{u}_z\right]\,. \tag{36}$$

This simple expression establishes a connection between the real part of the impedance Z and the imaginary part of the Green's function $\mathbf{G}$, or equivalently the LDOS.

5 Purcell factor

The change in the spontaneous decay rate Γ/Γ_0 induced by the environment of the emitter is known as the Purcell effect. Indeed, Purcell described this effect in a seminal paper [11], considering an emitter in a single mode cavity. In this section we show that the formalism described above includes Purcell's result.

The expansion (31) of the Green's function is valid in the absence of dissipation. For a cavity with weak losses, mode attenuation can be accounted for phenomenologically by introducing a damping rate γ_n for each mode, and rewriting the expansion as

$$\mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) = \sum_n c^2\,\frac{\mathbf{e}_n(\mathbf{r}) \otimes \mathbf{e}_n^*(\mathbf{r}')}{\omega_n^2 - \omega^2 - i\omega\gamma_n}\,. \tag{37}$$

It can be verified that (31) is recovered in the limit $\gamma_n \to 0$.

Following Purcell's approach, we assume that the emitter is resonant with one specific eigenmode ($\omega = \omega_n$) that dominates in the expansion of the Green's function. From Eqs. (13) and (37) we find that

$$\Gamma = \frac{2}{\epsilon_0 \hbar} |\mathbf{p}_{eg}|^2 \, Q \, |\mathbf{e}_n(\mathbf{r}_s) \cdot \mathbf{u}|^2 \tag{38}$$

where $\mathbf{u}$ defines the orientation of the transition dipole $\mathbf{p}_{eg}$ and $\mathbf{r}_s$ is the position of the emitter. We have introduced the quality factor of the mode $Q = \omega/\gamma_n$. The last term defines the so-called mode volume V such that

$$\frac{1}{V} = |\mathbf{e}_n(\mathbf{r}_s) \cdot \mathbf{u}|^2 , \tag{39}$$

which measures the volume occupied by the cavity mode. From Eqs. (14) and (38), we find that the normalized decay rate for a single mode cavity with weak losses reads

$$\frac{\Gamma}{\Gamma_0} = \frac{3}{4\pi^2} \lambda^3 \frac{Q}{V} \tag{40}$$

where $\lambda = 2\pi c/\omega$ is the mode wavelength. The right-hand side in the equation above is known as the Purcell factor. It describes the change in the LDOS induced by a single mode cavity.

References

[1] K. Drexhage, J. Lumin. **1-2**, 693 (1970); R. R. Chance, A. Prock and R. Silbey, Adv. Chem. Phys. **37**, 1 (1978).

[2] R. Carminati, A. Cazé, D. Cao, F. Peragut, V. Krachmalnicoff, R. Pierrat and Y. De Wilde, Surf. Sci. Rep. **70**, 1 (2015).

[3] D. Bouchet and R. Carminati, "Quantum dipole emitters in structured environments: a scattering approach (tutorial)", J. Opt. Soc. Am. A **36**, 186 (2019).

[4] R. Carminati and J.C. Schotland, *Principles of Scattering and Transport of Light* (Cambridge University Press, 2021).

[5] L. Novotny and B. Hecht, *Principles of Nano-Optics* (Cambridge University Press, 2006).

[6] G.S. Agarwal, Phys. Rev. A **11**, 230 (1975).

[7] J.M. Wylie and J.E. Sipe, Phys. Rev. A **30**, 1185 (1984).

[8] M. Born, *Atomic Physics* (Dover, New York, 1936), chap. V, sect. 8.

[9] R.J. Glauber and M. Lewenstein, Phys. Rev. A **43**, 467 (1991).

[10] J.-J. Greffet, M. Laroche and F. Marquier, Phys. Rev. Lett. **105**, 117701 (2010).

[11] E.M. Purcell, Phys. Rev. **69**, 681 (1946).

[12] S. Haroche, "Cavity Quantum Electrodynamics", in *Fundamental Systems in Quantum Optics*, J. Dalibard, J.M. Raimond and J. Zinn-Justin, eds, Les Houches Sesssion LIII, 1990 (Elsevier Science Publishers, 1992).