

# Tailoring spontaneous emission and scattering with antennas and cavities.

4

## 4.1 Spontaneous emission

The goal of this chapter is to discuss how the presence of a resonator modifies the spontaneous emission of a two-level system characterized by an electric dipolar transition. We will compare the classical antenna point of view and the quantum point of view for radiation emission. We will specifically discuss what is quantum and what is classical in the spontaneous emission process. We start by reviewing different points of view to analyse spontaneous emission in vacuum before analysing the role of an antenna.

### Decay rate of a classical oscillator - Larmor Formula

We start by establishing the connection between the power radiated in vacuum and the decay rate of the energy of the oscillator. We model the emitter using a harmonic oscillator with position  $x(t) = x_0(t) \cos(\omega_0 t)$ , where  $x_0(t)$  is a slowly varying amplitude, velocity  $v(t) = \dot{x}(t)$ , and electric dipole moment  $-ex(t)$ . The oscillator has a resonant frequency  $\omega_0$  and a mass  $m$ . The instantaneous energy is given by

$$U = \frac{1}{2}mv^2(t) + \frac{1}{2}m\omega_0^2x^2(t). \quad (4.1)$$

The amplitude  $x_0(t)$  is a slowly decaying function of time due to radiative losses. Assuming a slow amplitude decay  $\dot{x}_0 \ll \omega_0 x_0$ , we can approximate the energy by

$$U = \frac{1}{2}m\omega_0^2x_0^2(t). \quad (4.2)$$

We know compute explicitly the total power radiated by a point dipole. We start by using the retarded potential expression in the dipole approximation ; :

#### Vector potential of a point monochromatic dipole at frequency $\omega$

$$\tilde{\mathbf{A}}(\mathbf{r}) = \frac{\mu_0}{4\pi} \frac{e^{i\frac{\omega}{c}r}}{r} (-i\omega \mathbf{p}_0) \quad (4.3)$$

The dipole approximatio basically means that source currents reduces to a single point and no dephasing and retardation effects are to be expected due to the spatial extent of the source. We deduce directly the radiated electric field into the far field using plane wave relations between  $\mathbf{E}$  and  $\mathbf{A}$  :

$$\tilde{\mathbf{E}}(\mathbf{r}) = i\omega \tilde{\mathbf{A}}(\mathbf{r})_{\perp} \quad (4.4)$$

$$= \frac{\mu_0}{4\pi} \frac{e^{i\frac{\omega}{c}r}}{r} \omega^2 \mathbf{p}_{0,\perp} \quad \left( \propto \frac{\partial^2 \mathbf{p}_{0,\perp}}{\partial t^2} \right) \quad (4.5)$$

where the  $\perp$  label refers to the transverse component of a vector, or in other words, for a given direction of observation  $\mathbf{u}$ , the component of the vector which is perpendicular to  $\mathbf{u}$ .

We now use spherical coordinates and a dipole oriented along the  $\mathbf{u}_z$ , direction, so that  $|\mathbf{p}_{0\perp}| = |\mathbf{p}_0| \sin \theta$ .

The power  $dP$  radiated into a solid angle  $d\Omega$  is related to flux of the Poynting vector  $\langle \mathbf{S}_{\text{Poynting}} \rangle$  across an oriented element of surface,  $d\mathbf{\Sigma} = d\mathbf{\Sigma} \mathbf{u}_r$ , located far away from the dipole, so that the far field approximation is valid. In spherical coordinates, we can express  $d\mathbf{\Sigma}$  from the elements  $d\theta$  et  $d\varphi$  of polar and azimuthal angles :

$$d\mathbf{\Sigma} = r \sin \theta \, d\varphi \, r \, d\theta$$

The power radiated into a given plane wave direction is then :

$$\begin{aligned} \langle \mathbf{S}_{\text{Poynting}} \rangle \cdot d\mathbf{\Sigma} &= \langle S_{\text{Poynting}} \rangle \mathbf{u}_r \cdot d\mathbf{\Sigma} \mathbf{u}_r \\ &= I(\theta, \varphi) r^2 \sin \theta \, d\theta \, d\varphi \end{aligned}$$

where the radiometric intensity  $I(\theta, \varphi)$  is given by the time averaged value of the Poynting vector. For a single point dipole, we have :

### Radiation pattern of a dipole

$$\begin{aligned} I(\theta, \varphi, r) &= \frac{c \epsilon_0}{2} \omega^2 |\mathbf{A}(\mathbf{r})_{\perp}|^2 \\ &= \frac{\mu_0}{32\pi^2 c} \frac{\omega^4 |\mathbf{p}_{0\perp}|^2}{r^2} \\ &= \frac{\mu_0}{32\pi^2 c} \frac{\omega^4 |\mathbf{p}_0|^2 \sin^2 \theta}{r^2} \end{aligned}$$

A dipole radiated in the entire space, but not along its own direction. The radiated power is maximal in the perpendicular plane.

We get the total radiated power by integrating over angles  $\theta$  et  $\varphi$  :

### Total radiated power : Larmor's formula

$$\begin{aligned}
P_R &= \int_0^\pi \int_0^{2\pi} I(\theta, \varphi) r^2 \sin\theta \, d\theta \, d\varphi \\
&= \frac{\mu_0}{16\pi c} p_0^2 \int_0^\pi \sin^3\theta \, d\theta = \frac{1}{4\pi\epsilon_0} \frac{\omega^4 p_0^2}{3c^3}
\end{aligned}$$

From the previous expression, called Larmor's formula, we can derive a spontaneous decay time. We can write an energy-conservation equation where  $P_R$  stands for the Larmor formula of the power radiated by an electric dipole :

$$\frac{dU}{dt} = -P_R(t) = -\frac{\omega_0^4 e^2 x_0^2(t)}{12\pi\epsilon_0 c^3} = -\gamma_{\text{cl}} U. \quad (4.6)$$

### Classical spontaneous decay rate

The classical decay rate  $\gamma_{\text{cl}}$  is therefore given by

$$\gamma_{\text{cl}} = \frac{e^2 \omega_0^2}{6\pi\epsilon_0 c^3 m}. \quad (4.7)$$

As expected, the larger the dipole moment, the larger the radiated power and the decay rate.

### Quantum spontaneous-emission decay rate in vacuum

The full quantum calculation of the decay rate in vacuum gives a very different result with a different frequency dependence\* :

### Quantum spontaneous decay rate

$$\gamma_q = \frac{|d_{12}|^2 \omega^3}{3\pi\epsilon_0 c^3 \hbar}. \quad (19.5)$$

Here  $d_{12}$  is the matrix element of the dipole operator and  $2\pi\hbar$  is Planck's constant. The presence of  $\hbar$  stresses the quantum nature of this result. It can be shown using the Wigner–Weisskopf method that the decay is exponential.

The decay rate of this exponential can be found using the Fermi Golden Rule :

$$\gamma_0 = \frac{2\pi}{\hbar^2} |\hat{W}_{if}|^2 g(\omega), \quad (4.8)$$

\* The full derivation can be found in reference textbooks such as Loudon, Quantum Theory of Light.

where  $\hat{W}_{if} = \langle f | \hat{\mathbf{d}} \cdot \hat{\mathbf{E}} | i \rangle$  is the matrix element of the interaction Hamiltonian between the initial and the final states, and  $g(\omega)d\omega$  is the number of electromagnetic states in the frequency interval  $d\omega$  with the electric field parallel to the dipole moment. Let us stress that an electromagnetic state is nothing but a mode of Maxwell's equations (e.g. a mode in a cavity or a waveguide).

In vacuum, the states or modes are plane waves characterized by a frequency and a wavevector. The projected density of states (DOS) is given by :

$$g(\omega) = V \frac{\omega^2}{3\pi^2 c^3}. \quad (4.9)$$

It can be found by applying boundary conditions on a cubic volume  $V = L^3$  containing the electric field to count modes in k-space, using the dispersion relation in vacuum, and finally a 1/3 factor to get the density of states in a given polarization orientation.

Using the quantized electromagnetic field with amplitude  $\sqrt{\hbar\omega/2\epsilon_0 V}$ , it is found that the interaction Hamiltonian matrix element is

$$|\hat{W}_{if}|^2 = \frac{|d_{12}|^2 \hbar\omega}{2\epsilon_0 V}. \quad (4.10)$$

Upon using the previous expressions, we obtain the radiative decay rate in vacuum given at the beginning of this section.

Although the quantum result is very different from the classical one, it is interesting to note that it is possible to recover the quantum result by simply replacing two terms in the classical calculation by their quantum counterpart. The first term is the classical energy

$$U = \frac{1}{2} m \omega_0^2 x_0^2, \quad (4.11)$$

which can be replaced by  $\hbar\omega$ , and the second term is the classical dipole moment  $ex_0$ , which can be replaced by  $2d_{12}$ .

## Classical radiation in vacuum : Radiation reaction work

In this section, we establish a connection between the power radiated and the power exchanged between the dipole and the radiation reaction field, namely the field radiated by a source on itself. We start by considering the power radiated by a classical dipole in vacuum. We consider a dipole source inside a volume  $V$  enclosed by a surface  $A$ . The integral form of the energy conservation equation yields

$$\int_V \frac{\partial u}{\partial t} d^3r = - \int_V \mathbf{j}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}) d^3r - \int_A \mathbf{S} \cdot d\mathbf{A}, \quad (4.12)$$

where  $u$  is the electromagnetic energy density,  $\mathbf{j}$  is the current density, and  $\mathbf{E}$  is the electric field.

We now consider a monochromatic point-like dipole source with dipole moment  $\mathbf{p}$  in stationary regime such that  $\mathbf{j}(\mathbf{r}) = -i\omega\mathbf{p}\delta(\mathbf{r} - \mathbf{r}')$ . We can compute the time-averaged energy-conservation relation using complex amplitudes :

$$\frac{1}{2}\Re[i\omega\mathbf{p}\cdot\mathbf{E}^*] = \int_A \langle \mathbf{S} \rangle \cdot d\mathbf{A} = P_R, \quad (4.13)$$

where  $P_R$  is the radiated power. The term on the left-hand side involves the electromagnetic field generated by the dipole at its own position, namely the radiation reaction.

The electric field generated by the dipole source can be cast in the form

$$\mathbf{E}(\mathbf{r}) = \mu_0\omega^2 \overleftrightarrow{\mathbf{G}}^{(0)}(\mathbf{r}, \mathbf{r}')\mathbf{p}, \quad (4.14)$$

where we have introduced the vacuum Green tensor :

$$\overleftrightarrow{\mathbf{G}}^{(0)}(\mathbf{r}, \mathbf{r}') = \text{PV} \left[ \overleftrightarrow{\mathbf{I}} + \frac{1}{k_0^2} \nabla \nabla \right] \frac{e^{ik_0 R}}{4\pi R} - \frac{\overleftrightarrow{\mathbf{I}}}{3k_0^2} \delta(\mathbf{r} - \mathbf{r}'), \quad (19.11)$$

where PV stands for the principal value,  $R = |\mathbf{r} - \mathbf{r}'|$ , and  $k_0 = \omega/c$ .

Using the Green tensor relation to the electric field, the power generated by a dipole  $\mathbf{p} = p_z \mathbf{e}_z$  can be written as

$$P_R^{(0)} = \frac{\mu_0\omega^3}{2} \Im \left[ G_{zz}^{(0)}(\mathbf{r}, \mathbf{r}) \right] |p_z|^2. \quad (4.15)$$

Although the real part of  $G_{zz}^{(0)}$  diverges, its imaginary part is well defined and takes the value  $\omega/(6\pi c)$  (to be derived in detail in appendices...). We thus recover the usual Larmor formula for the power radiated by a dipole :

$$P_R^{(0)} = \frac{\mu_0\omega^3}{2} \frac{\omega}{6\pi c} |p_z|^2 = \frac{1}{4\pi\epsilon_0} \frac{\omega^4 |\mathbf{p}|^2}{3c^3}. \quad (4.16)$$

## Classical radiation in the presence of a resonator

We now consider the power radiated by a classical dipole source with dipole moment  $\mathbf{p}$  in the presence of a resonator. The resonator can be any object made of an arbitrary material : a cavity, a nanoantenna, or a complex environment. The dipole and the resonator are located in a volume  $V$ . The dipole drives a current density  $\mathbf{j}_{\text{ant}}(\mathbf{r}')$  in the resonator, leading to radiation and to a material dissipation rate  $\mathbf{j}_{\text{ant}}(\mathbf{r}') \cdot \mathbf{E}(\mathbf{r}')$ , where  $\mathbf{E}(\mathbf{r}')$  is the induced electric field.

Repeating the analysis of the previous section, we find a decay rate given by the Green tensor  $G_{zz}$  instead of  $G_{zz}^{(0)}$ . Energy conservation can now be written as

$$\frac{1}{2}\Re[i\omega\mathbf{p}\cdot\mathbf{E}^*] = \int_V \mathbf{j}_{\text{ant}}(\mathbf{r}) \cdot \mathbf{E}(\mathbf{r}) d^3r + \int_A \mathbf{S} \cdot d\mathbf{A}. \quad (4.17)$$

This equation shows that the power delivered by the dipole equals the sum of the radiated power  $P_R$  and a non-radiative power dissipated in the resonator,  $P_{NR}$ . Hence,

$$\frac{\mu_0\omega^3}{2} \Im[G_{zz}(\mathbf{r}, \mathbf{r})] |\mathbf{p}|^2 = P_{NR} + P_R. \quad (4.18)$$

The Green tensor now provides the field produced by the dipole in the presence of the resonator :

$$\mathbf{E}(\mathbf{r}) = \mu_0\omega^2 \overleftrightarrow{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \mathbf{p}. \quad (19.20)$$

The radiation reaction thus includes the field radiated by the resonator back onto the dipole source. The only formal modification with respect to vacuum is the replacement  $\overleftrightarrow{\mathbf{G}}^{(0)} \rightarrow \overleftrightarrow{\mathbf{G}}$ . The decay rate is therefore proportional to  $\Im[G_{zz}]$ .

The decay rate  $\gamma_{\text{ant}}$  in the presence of the antenna is given by

$$\frac{\gamma_{\text{ant}}}{\gamma_0} = \frac{\Im[G_{zz}]}{\Im[G_{zz}^{(0)}]} = \frac{P_R + P_{NR}}{P_R^{(0)}}. \quad (19.21)$$

Although the spontaneous-emission decay rate depends on  $\hbar$  and on the quantum electric-dipole-moment matrix element, the decay rate in an arbitrary environment is simply the vacuum decay rate multiplied by a correction factor accounting for the modification of the radiation reaction.

From a quantum point of view, this correction can be interpreted as a modification of the density of states. Since the decay rate depends on the exact position of the emitter, this requires the use of the local density of states (LDOS)  $\rho(\mathbf{r}, \omega)$ . The LDOS accounts for the spatial structure of the modes, which exhibit nodes and antinodes. The coupling strength depends on the overlap between the dipole moment and the local mode field.

Finally, the decay rate can be enhanced either by an increase of the radiated power or by additional decay channels due to losses in the resonator. The LDOS derived from the imaginary part of the Green tensor naturally includes non-radiative decay channels, which are negligible in dielectric or microwave cavities but play a crucial role in plasmonic nanoantennas.

## When quantum physics meets classical physics

We now compare the classical and quantum approaches. The connection between the two points of view can be established in a simple manner. In vacuum, we have

$$\frac{\omega^2}{\pi^2 c^3} = \frac{6\omega}{\pi c^2} \text{Im} \left[ G_{zz}^{(0)}(\mathbf{r}, \mathbf{r}') \right], \quad (4.19)$$

where the term on the left-hand side was introduced as a density of electromagnetic states, while the term on the right-hand side was introduced as the radiation reaction.

If we now consider a more general situation than vacuum by introducing resonators, the radiation reaction is still well defined.

The local density of electromagnetic states (LDOS)  $\rho(\mathbf{r},\omega)$  is proportional to the imaginary part of the trace of the Green tensor :

$$\rho(\mathbf{r},\omega) = \frac{2\omega}{\pi c^2} \operatorname{Im} \left[ \operatorname{tr} \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}) \right]. \quad (4.20)$$

This expression is a general formula for the LDOS in the presence of lossy objects. When dealing with spontaneous emission, we restrict the LDOS to the axis parallel to the dipole moment, leading to the projected LDOS. Inserting this form into the spontaneous-emission decay rate, we obtain

$$\gamma_R = \frac{2\pi}{\hbar^2} |\hat{W}_{if}|^2 \frac{2\omega}{\pi c^2} \operatorname{Im}[G_{zz}(\mathbf{r},\mathbf{r})]. \quad (4.21)$$

It is seen that the structure of the decay rate is given by the product of a quantum term,  $(2\pi/\hbar^2) |\hat{W}_{if}|^2$ , and the LDOS, which is a purely classical quantity. By comparing the decay rate in the presence of an antenna with that in vacuum, we recover the simple relation

$$\gamma_{\text{ant}} = \gamma_0 \frac{\operatorname{Im}[G_{zz}(\mathbf{r},\mathbf{r})]}{\operatorname{Im}[G_{zz}^{(0)}(\mathbf{r},\mathbf{r})]}. \quad (4.22)$$

To summarize, the classical point of view identifies the power radiated with the power associated with radiation reaction, which is proportional to  $\operatorname{Im}[G_{zz}]$ . The quantum point of view shows that the decay rate is proportional to the density of states, which is itself proportional to  $\operatorname{Im}[G_{zz}]$ . Thus, modifying the environment of an emitter amounts to modifying both the LDOS and the radiation reaction, which are two sides of the same physical phenomenon. As a result, a classical calculation of the LDOS modification allows the spontaneous emission rate to be predicted.

### Controlling $\operatorname{Im}[G_{zz}]$ with a cavity : LDOS and the Purcell factor

The first proposal to control spontaneous emission by modifying the electromagnetic environment was introduced by Purcell in the context of nuclear magnetic resonance in the microwave regime. We now give a back-of-the-envelope derivation of the modification of the spontaneous emission rate by a multiplicative factor  $F_P$ , known as the Purcell factor.

We consider an emitter placed inside a single-mode cavity characterized by a mode volume  $V$  and a decay rate  $\kappa$ . Although the cavity supports a single mode, radiative and non-radiative losses broaden its spectrum. If the cavity linewidth  $\kappa$  is much larger than the intrinsic linewidth  $\gamma_R$  of the two-level system, the cavity can be treated as a quasi-continuum, and the Fermi Golden Rule applies.

Assuming a Lorentzian cavity resonance, the normalized spectral density is

$$g(\omega) = \frac{1}{2\pi} \frac{\kappa}{(\omega - \omega_0)^2 + \kappa^2/4}. \quad (4.23)$$

At resonance, the spectral density takes the value  $g(\omega_0) = 2/(\pi\kappa)$ . Since there is a single mode in a volume  $V$ , and assuming a uniform field distribution for simplicity, the local density of states is given by  $g(\omega_0)/V$ .

The enhancement factor of the decay rate, also called the Purcell factor  $F_P = \gamma_{\text{cav}}/\gamma_R$ , is given by the ratio of the LDOS in the cavity to that in vacuum :

$$F_P = \frac{\gamma_{\text{cav}}}{\gamma_R} = \frac{2}{\pi\kappa V} \frac{3\pi^2 c^3}{\omega^2 n^3} = \frac{3Q}{4\pi^2} \frac{\lambda^3}{V}, \quad (4.24)$$

where  $2\pi/\lambda = n\omega/c$  and the cavity quality factor is defined as  $Q = \omega/\kappa$ .

This is the form originally introduced by Purcell. The Purcell factor quantifies both spatial confinement, through the ratio  $\lambda^3/V$ , and spectral confinement, through the quality factor  $Q$ . Dielectric microcavities can exhibit very high quality factors ( $Q > 10^5$ ), but typically have moderate spatial confinement. Conversely, plasmonic resonators provide extreme spatial confinement at the expense of lower  $Q$ -factors.

## 4.2 Light source engineering using fluctuational electrodynamics and Kirchhoff's law

### The fluctuation electrodynamics approach

We recall that, for a given system, the field generated by an arbitrary distribution of monochromatic sources is expressed as

$$\mathbf{E}(\mathbf{r},\omega) = i\omega\mu_0 \int_V \overleftrightarrow{\mathbf{G}}(\mathbf{r},\mathbf{r}',\omega) \mathbf{j}(\mathbf{r}',\omega) d^3\mathbf{r}', \quad (4.25)$$

where  $V$  denotes the source volume.

The central quantity underlying all radiometric observables of a light-emitting system is the radiated power, i.e. the Poynting vector  $\mathbf{\Pi}(\mathbf{r},\omega)$ . Since we are dealing with spontaneous emission—a macroscopic manifestation of stochastic processes occurring at the microscopic scale—the fields must be treated as random processes, driven by random currents. Accordingly, the Poynting vector depends on the statistical average of the squared modulus of the electric field. In the far-field limit, the intensity radiated in the direction  $\mathbf{u} = \mathbf{r}/r$  can be written as

$$I(\mathbf{u},\omega) = |\mathbf{\Pi}(\mathbf{r},\omega)| = c\epsilon_0(\omega^2\mu_0^2)\langle|\mathbf{E}(\mathbf{r},\omega)|^2\rangle = \sum_{k,l,m} \int_V d^3\mathbf{r}_1 \int_V d^3\mathbf{r}_2 G_{kl}(\mathbf{r}_1, \mathbf{r}_2) G_{km}^*(\mathbf{r}_1, \mathbf{r}_2) \langle j_l(\mathbf{r}_1, \omega) j_m^*(\mathbf{r}_2, \omega) \rangle \quad (4.26)$$

where angular brackets denote statistical averaging, and indices  $k,l,m$  correspond to the  $x,y,z$  Cartesian coordinates.

The classical interpretation is straightforward : random charge motion in the active medium induces random currents. Since the mean of these currents is zero, the average radiated field also vanishes. However, because radiated power is *quadratic* in the field, its mean value

is non-zero. The relevant source term thus emerges as  $\langle j_l(\mathbf{r}_1, \omega) j_m^*(\mathbf{r}_2, \omega) \rangle$ , i.e. the current density correlation function. In other words, spontaneous emission—being intrinsically stochastic—can be interpreted as radiation originating from current fluctuations in the active medium. This constitutes the foundation of fluctuational electrodynamics, as introduced by Sergueï Rytov starting in the 1950s.

Equation (4.26) can be intuitively interpreted term by term. The Green's tensor encodes the electromagnetic environment : it defines the system's mode structure and quantifies how modes couple to the far field when driven by sources. It is a purely classical quantity. By contrast, the current correlation function carries microscopic quantum information, as it reflects the dynamics of elementary charges in the electronic states of the active medium—namely, their energy levels and population distributions. Taken together, the full expression describes light emission as the response of an ensemble of electromagnetic modes, driven by microscopic excitations, and leaking into the far field.

At first glance, this excitation–response formulation seems close to the local point-dipole approach. However, the latter requires explicitly defining a distribution of sources—positions, orientations, amplitudes—which is intrinsically inadequate to describe ensembles of interacting emitters. Worse still, it often relies on arbitrary parameters.

By contrast, the fluctuational approach admits analytical source terms under a very limited set of assumptions. The essential requirement is that the active medium reaches local thermodynamic equilibrium, a “thermalization condition.” Under this condition, electronic excitations can be described using the tools of quantum statistical mechanics, with only a few thermodynamic parameters such as temperature  $T$  and chemical potential  $\mu$ . This establishes a strong connection to condensed-matter physics, where statistical methods efficiently capture macroscopic behavior emerging from large ensembles of quantum systems.

The key insight here is that the current correlation function—the source term—can be explicitly determined in many practical cases, either through the fluctuation–dissipation theorem or its extensions. As a result, the radiated power of most luminescent systems can be calculated in absolute terms, without recourse to arbitrary parametrization.

In the following, we outline the essential steps of this discussion.

## From Fluctuation-Dissipation Theorem and the equilibrium picture to incandescent light emission

Since random currents cannot be represented by square-integrable functions, it is convenient to describe them in terms of power density. In this framework, the current correlation function takes the form

$$\langle j_l(\mathbf{r}_1, \omega) j_m^*(\mathbf{r}_2, \omega') \rangle = 2\pi\delta(\omega - \omega') W_{j_l j_m}(\mathbf{r}_1, \mathbf{r}_2, \omega), \quad (4.27)$$

where  $W_{j_l j_m}(\mathbf{r}_1, \mathbf{r}_2, \omega)$  is the cross-spectral density.

Let us now assume that the system is homogeneous and in thermodynamic equilibrium at temperature  $T$ . In this case, current fluctuations are purely thermal. Using the quantum form of the fluctuation–dissipation theorem, one obtains

$$W_{j_1 j_m}(\mathbf{r}_1, \mathbf{r}_2, \omega) = 2\omega\epsilon_0 \operatorname{Im}[\epsilon(\omega)] \left[ \frac{\hbar\omega}{\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1} \right] \delta_{lm} \delta(\mathbf{r}_1 - \mathbf{r}_2), \quad (4.28)$$

where the zero-point contribution has been neglected.

We previously emphasized that the current correlation—or equivalently, the cross-spectral density—characterizes the source term that excites the electromagnetic modes of the system. Equation (4.28) makes this role explicit :

- The factor  $\operatorname{Im}[\epsilon(\omega)]$  represents the dissipative part of the material response, i.e. how efficiently matter absorbs light. It vanishes at frequencies  $\omega$  where no optical transitions are available, and becomes large where many transitions occur. Physically, it acts as a light–matter coupling coefficient, linked to transition matrix elements and to the optical joint density of states.
- The term  $\left[ \frac{\hbar\omega}{\exp(\hbar\omega/k_B T) - 1} \right]$  corresponds to the Bose–Einstein distribution multiplied by the photon energy  $\hbar\omega$ . It describes the occupation of optically active states by excitation quanta, i.e. the spectral distribution of available energy in the medium under thermal equilibrium.

By inserting this source term into the expression for the radiated power, one recovers Planck's law of blackbody radiation.

This analysis of the cross-spectral density also provides a more general conceptual framework. Beyond strict thermal equilibrium, one can still construct effective cross-spectral densities that describe emission from non-thermal systems. The idea is that photons form a Bose gas in equilibrium (or quasi-equilibrium) with a reservoir of electronic excitations. Their energy distribution can thus be described by the same thermodynamic parameters—temperature  $T$  and chemical potential  $\mu$ —that characterize the electronic system, whenever such quantities are well-defined.

In this picture, light emission corresponds to the occupation of radiation modes by photons, mediated by both the system's permittivity  $\epsilon(\omega)$  and the Green's tensor that encodes the electromagnetic environment.

As a limiting case, Planck's law corresponds to a situation where the excitation reservoir follows a thermal distribution, defined by a uniform temperature  $T$  in a homogeneous bulk medium.

## Radiation by non-equilibrium, but thermalized systems : example of a semiconductor

Let us now consider a different case : a semiconductor under electrical or optical pumping.

Semiconductors act as large reservoirs of electrons and holes. At thermodynamic equilibrium, the conduction electrons follow a Fermi-Dirac distribution at temperature  $T$  with Fermi level  $\mu_F$ , a condition maintained by recurrent interactions among electrons and with the lattice (the thermostat). In intrinsic semiconductors, the corresponding distribution of valence holes follows directly, since it mirrors the distribution of electrons. Thus, the excitation distribution is thermal, defined by the single parameter  $T$ . In this case, the material emits thermal radiation, but with very low intensity, since conduction-band states are only sparsely occupied at room temperature.

Now, let us shine an intense femtosecond laser pulse with energy above the band gap. A substantial population of electrons is promoted to the conduction band, leaving holes in the valence band. Immediately after excitation, the carriers are far from equilibrium. However, within a few hundred femtoseconds, strong electron-electron and electron-phonon scattering establishes a local thermodynamic equilibrium separately within the conduction and valence bands. In contrast, radiative recombination occurs on nanosecond time scales, much slower. As a result, the electron and hole populations can be described by independent Fermi-Dirac distributions, characterized by a common carrier temperature  $T$  and distinct quasi-Fermi levels  $\mu_c$  and  $\mu_v$  :

$$f_c^{\text{FD}}(E) = \frac{1}{\exp\left(\frac{E-\mu_c}{k_B T}\right) + 1}, \quad f_v^{\text{FD}}(E) = \frac{1}{\exp\left(\frac{E-\mu_v}{k_B T}\right) + 1}. \quad (4.29)$$

The probability of emitting a photon of energy  $\hbar\omega$  is proportional to the joint probability that an electronic state in the conduction band is occupied while the corresponding valence-band state is empty. For a transition between joint states at energies  $E$  and  $E + \hbar\omega$ , one has

$$\eta_{\text{sp}}(\omega) \propto f_c^{\text{FD}}(E + \hbar\omega)[1 - f_v^{\text{FD}}(E)], \quad (4.30)$$

the proportionality factor including the Einstein  $A$  coefficient and the joint density of states.

This relation can be connected to the absorption coefficient of the material, which itself depends on the carrier distributions. Indeed, the net absorption is reduced as conduction-band states fill, leading to state-blocking and, under pumping, to transparency or even gain. Accounting for both stimulated absorption and stimulated emission, the absorption coefficient of the material reads

$$\alpha(\omega, \mathbf{r}) \propto f_v^{\text{FD}}(E)[1 - f_c^{\text{FD}}(E + \hbar\omega)] - f_c^{\text{FD}}(E + \hbar\omega)[1 - f_v^{\text{FD}}(E)] = f_v^{\text{FD}}(E) - f_c^{\text{FD}}(E + \hbar\omega). \quad (4.31)$$

From these relations, the ratio of spontaneous emission to net absorption can be computed in closed form :

### Universal luminescence-absorption relation

$$\frac{\eta_{\text{sp}}(\omega)}{\alpha(\omega)} \propto \frac{1}{\exp\left(\frac{\hbar\omega-\mu}{k_B T}\right) - 1}, \quad \mu = \mu_c - \mu_v, \quad (4.32)$$

where  $\mu$  is the difference in quasi-Fermi levels. As highlighted by Gordon Lasher and Frank Stern, then shortly later by Boris Steapnov and Victor Gribkovskii, Equation (4.32) is a universal relation between spontaneous emission and absorption coefficient whenever local thermodynamic equilibrium is established.

Rewriting (4.32) highlights the analogy with the cross-spectral density in Eq. (4.28) :

$$f_c^{\text{FD}}(E + \hbar\omega)[1 - f_v^{\text{FD}}(E)] \propto \alpha(\omega) \frac{\hbar\omega}{\exp\left(\frac{\hbar\omega-\mu}{k_B T}\right) - 1}. \quad (4.33)$$

This form makes the physics transparent : spontaneous emission appears as the product of

1. a dissipation term, the absorption coefficient  $\alpha(\omega)$  (proportional to  $\text{Im}[\epsilon(\omega)]$ ), and
2. a Bose-Einstein factor, now with a non-zero chemical potential  $\mu$ .

Thus, under pumping – and provided thermalization within each band is reached – the electronic excitations form populations described by equilibrium statistical distributions characterized by  $T$  and  $\mu$ . Light emission can then be viewed as a Bose gas of photons in equilibrium with electronic excitations. The chemical potential  $\mu$ , introduced and popularized by Würfel as the chemical potential of radiation, is directly given by the quasi-Fermi level separation. It therefore quantifies the pumping strength : the stronger the pumping, the larger the separation  $\mu_c - \mu_v$ , and the greater the chemical potential of the emitted photons.

### Elements of a generalized theory of luminescence

The previous example, based on semiconductors, enabled us to explicitly introduce distribution functions and connect luminescence to well-known statistical arguments. When discussing luminescence processes, it is often tempting to draw sharp distinctions between incandescence, photoluminescence, electroluminescence, and other mechanisms. Yet, the fundamental difference lies only in the pumping mechanism used to populate the excited states. The emission process itself is always the radiative decay of excited states into lower-energy states.

This observation highlights the scope of the present framework : it applies as soon as excited and ground state populations can be defined, together with a dissipation coefficient, and provided that thermalization occurs faster than radiative decay. In such cases, the excited-state populations can be described by thermodynamic quantities, which can even be defined locally through a temperature field  $T(\mathbf{r})$  and a chemical potential field  $\mu(\mathbf{r})$ .

With this in mind, we can extend fluctuational electrodynamics to describe photoluminescence and electroluminescence. The only modification is the generalization of the source term in the cross-spectral density :

$$W_{j_l j_m}(\mathbf{r}, \mathbf{r}', \omega) = 2\omega\epsilon_0 \operatorname{Im}[\epsilon(\mathbf{r}, \omega)] \left[ \frac{\hbar\omega}{\exp\left(\frac{\hbar\omega - \mu(\mathbf{r})}{k_B T(\mathbf{r})}\right) - 1} \right] \delta_{lm} \delta(\mathbf{r}_1 - \mathbf{r}_2), \quad (4.34)$$

where  $\operatorname{Im}[\epsilon(\mathbf{r}, \omega)]$  accounts for the dissipative response of the pumped system.

From this expression, the power radiated by a volume element  $d^3\mathbf{r}'$  around position  $\mathbf{r}'$  follows as

$$|\mathbf{\Pi}(\mathbf{r}, \omega)| \propto 4\pi \frac{\omega^3}{c^3} \sum_{k,l,m} \int_V d^3\mathbf{r}' |G_{kl}(\mathbf{r}, \mathbf{r}')|^2 \operatorname{Im}[\epsilon(\mathbf{r}', \omega)] \left[ \frac{\hbar\omega}{\exp\left(\frac{\hbar\omega - \mu(\mathbf{r}')}{k_B T(\mathbf{r}')} \right) - 1} \right], \quad (4.35)$$

which can be recast as

### Generalized Planck's law

$$dP_{\text{rad}}^{(l)}(\mathbf{u}, \omega) = \int_V \eta^{(l)}(\mathbf{u}, \mathbf{r}', \omega) \frac{I_{\text{bb}}(T(\mathbf{r}'), \mu(\mathbf{r}'), \omega)}{2} d^3\mathbf{r}' d\Omega \quad (4.36)$$

where

$$I_{\text{bb}}(T(\mathbf{r}'), \mu(\mathbf{r}'), \omega) = \frac{\omega^2}{4\pi^3 c^2} \frac{\hbar\omega}{\exp\left(\frac{\hbar\omega - \mu(\mathbf{r}')}{k_B T(\mathbf{r}')} \right) - 1} \quad (4.37)$$

is a generalized blackbody radiance, defined locally by  $T(\mathbf{r}')$  and  $\mu(\mathbf{r}')$ .

Equation (4.36) therefore constitutes a generalized Planck's law of radiation, obtained directly from the principles of fluctuational electrodynamics in a local form.

### Computing emission from absorption : Kirchhoff's law

The only obstacle that remains in computing the light emission of an arbitrary thermalized system is the evaluation of the term  $\eta^{(l)}(\mathbf{u}, \mathbf{r}', \omega)$ , which we have identified as the emissivity density. From Eq. (4.35), we see that this quantity involves the product of the Green's tensor with the material dissipation term,

$$|G_{kl}(\mathbf{r}, \mathbf{r}_1)|^2 \operatorname{Im}[\epsilon(\mathbf{r}_1, \omega)]$$

and thus describes how the electromagnetic structure mediates the coupling of material excitations into radiation.

At first sight, this seems to require a forward calculation of emission from point-like sources distributed throughout the medium—a prohibitively heavy computational task. Fortunately, reciprocity principles allow us to reformulate the problem in terms of absorption, which can be evaluated at much lower cost. This is the essence of the generalized Kirchhoff's law. I rephrase below the major steps of its derivation.

### Absorption picture

Let us consider the reciprocal scenario : instead of fluctuating currents radiating outward, we illuminate the same medium with an incoming field (for instance, radiated by a point dipole located at  $\mathbf{r}$ ). The absorbed power in a small volume element  $d^3\mathbf{r}'$  is

$$dP_{\text{abs}}(\mathbf{r}') = \text{Im}[\epsilon(\mathbf{r}', \omega)] \frac{\omega\epsilon_0}{2} |\mathbf{E}(\omega, \mathbf{r}')|^2 d^3\mathbf{r}' \quad (4.38)$$

where  $\mathbf{E}(\omega, \mathbf{r}')$  is the local electric field generated by the incident source.

Using the Green's tensor to connect the source to the local field, we can rewrite this absorbed power as

$$dP_{\text{abs}} \propto |G_{kl}(\mathbf{r}', \mathbf{r})|^2 \text{Im}[\epsilon(\mathbf{r}', \omega)] \frac{\omega\epsilon_0}{2} |E_{\text{inc}}|^2 d^3\mathbf{r}' \quad (4.39)$$

This motivates the definition of the absorption cross-section density

$$\alpha(\mathbf{u}, \mathbf{r}', \omega) = |G_{kl}(\mathbf{r}', \mathbf{r})|^2 \text{Im}[\epsilon(\mathbf{r}', \omega)] \quad (4.40)$$

so that

$$dP_{\text{abs}} \propto \alpha(\mathbf{u}, \mathbf{r}', \omega) \frac{\omega\epsilon_0}{2} |E_{\text{inc}}|^2 d^3\mathbf{r}' \quad (4.41)$$

### Reciprocity and Kirchhoff's law

If the material is reciprocal, Lorentz reciprocity applies : the field generated at  $\mathbf{r}$  by a dipole at  $\mathbf{r}'$  is equal to the field generated at  $\mathbf{r}'$  by the same dipole placed at  $\mathbf{r}$ . In tensor form,

$$G_{kl}(\mathbf{r}, \mathbf{r}') = G_{lk}(\mathbf{r}', \mathbf{r}). \quad (4.42)$$

Inspection of Eqs. (4.35) and (4.39) then reveals that both emission and absorption involve the same tensorial quantity. As a result, we obtain the generalized Kirchhoff's law in local form :

$$\eta^{(l)}(\mathbf{u}, \mathbf{r}', \omega) = \alpha(\mathbf{u}, \mathbf{r}', \omega). \quad (4.43)$$

Physically, this means that the emissivity density is nothing but the normalized absorption efficiency of the medium. More explicitly,

$$\eta^{(l)}(\mathbf{u}, \mathbf{r}', \omega) = \frac{\text{Im}[\epsilon(\mathbf{r}', \omega)] \frac{\omega \epsilon_0}{2} |\mathbf{E}(\omega, \mathbf{r}')|^2 d^3 \mathbf{r}'}{\frac{\epsilon_0 c}{2} |E_{\text{inc}}|^2 d^3 \mathbf{r}'} \quad (4.44)$$

This dimensionless quantity is simply the ratio between the absorbed power in  $d^3 \mathbf{r}'$  and the power delivered by the incident wave within the same volume.

Substituting into Eq. (4.36), we arrive at the generalized Kirchhoff's law for emission :

### Generalized Kirchhoff's law

$$dP_{\text{rad}}^{(l)}(\mathbf{u}, \omega) = \int_V \alpha^{(l)}(\mathbf{u}, \mathbf{r}', \omega) \frac{I_{\text{bb}}(T(\mathbf{r}'), \mu(\mathbf{r}'), \omega)}{2} d^3 \mathbf{r}' d\Omega. \quad (4.45)$$

In summary : the forward emission problem (summing over dipole sources and evaluating Green's tensors everywhere) can be replaced by the reciprocal absorption problem (evaluating local absorption under plane-wave illumination). The generalized Kirchhoff's law ensures equivalence, while offering a numerically much simpler route whenever the active medium can be described by a dielectric function and thermodynamic parameters.

Most importantly, we highlight that Kirchhoff's law establishes a *detailed balance* between absorptivity and emissivity : emission by an active region of a system *in a given direction, at a given frequency, and in a given polarization state*, is equivalent to the fraction of absorption by the very same active region when the whole system is illuminated by a plane wave, coming *from the same given direction, at the same given frequency, and in the same polarization state*.

To design a source with desired features—angular pattern, spectrum, or polarization—the task reduces to optimizing the corresponding absorptivity. A spectrally narrow emitter must strongly absorb in a narrow frequency window ; a polarized source must absorb preferentially in one polarization ; a directional source must absorb preferentially from one incidence angle. This description applies to thermal emission, but also to other luminescence processes, as long as they can be described by equilibrium like distributions defined by a photon chemical potential.