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Spontaneous emission in absorbing dielectrics: an alternative approach

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Abstract

We consider the spontaneous emission in an absorbing medium by calculating the quantum flow of energy from an emitting atom. For this purpose, we extend the analysis by Power and Thirunamachandran of quantized Maxwell fields in the vicinity of an atom in the free space to a situation where the emitter is in a dielectric medium. We represent the quantum flow in terms of the non-equal time commutators involving electric and magnetic fields, as well as the local displacement field. The commutators are calculated applying a previously developed microscopic theory of quantization of radiation in dielectrics. Consequently, we derive from first principles the rate of spontaneous emission in an absorbing medium containing local field factors and other refractive contributions. The results support the previous introduction of the (virtual cavity) local field correction by Barnett *et al* on a phenomenological basis, as well as our previous approach in which the spontaneous emission is considered as a resonance energy transfer to the far-zone atoms. A distinctive feature of the present formalism is that it deals with the spontaneous emission directly by calculating the quantum flow from the emitting atom. Therefore, the method is free from divergences appearing in other approaches analysing the spontaneous decay in absorbing media.

This paper is dedicated to the memory of Edwin Power.

1. Introduction

During the past years, there has been a great deal of interest in modified spontaneous emission by atoms (molecules) in various environments, such as in photonic band-gap crystals [1–5] and left-handed media [6, 7]. Spontaneous emission has been investigated in ordinary

homogeneous dielectrics as well, including both transparent [8–11] and absorbing [12–18] ones. In the case of transparent dielectrics, the rate of the spontaneous emission is usually obtained through a standard procedure that involves summation over the modes of emitted photons [10, 11]. The rate of the spontaneous decay can then be represented in terms of the appropriate (single-site) Green functions. However, such a procedure leads to spurious divergences in the rate of the spontaneous decay if applied to absorbing media [13, 16, 17]. The divergences appear because the calculated rate of the spontaneous decay accommodates not only emission of a real photon but also de-excitation of the emitter by near-zone atoms via a Förster mechanism [19–22]. The Förster energy transfer represents an exchange of excitation energy between an excited and unexcited atom with a rate proportional to R^{-6}, where R is an inter-atomic separation. The decay rate of an excited atom is thus sensitive to its local atomic environment and diverges if a continuous limit is applied for the surrounding medium. The divergences can be removed by applying a real cavity model [23, 24] or by using other procedures that effectively introduce a cut-off distance preventing the surrounding atoms from being too close to the emitter [13, 16, 17]. It is worth noting that various procedures lead to somewhat different emission rates [13, 16, 17].

It is desirable to have a method allowing one to calculate the rate of spontaneous emission directly without eliminating the near-zone (Förster) energy transfer. In the previous paper [14], we have applied a unified theory of radiative and radiationless energy transfer in dielectrics [22] to deal with the spontaneous emission in absorbing media. The decay rate of an excited atom has been derived calculating the total rate of excitation transfer to the surrounding species. The contribution due to the far-zone energy transfer was shown to yield the rate of a photon emission in the absorbing medium [14].

Here, we develop a more direct method of calculating the rate of spontaneous emission in an absorbing medium. Specifically, we derive the emission rate in the absorbing medium by calculating the quantum flow of energy by an emitting atom. For this purpose, we extend the analysis by Power and Thirunamachandran of quantized Maxwell fields in a vicinity of an atom in the free space [25, 26] to a situation where the emitter is situated in a dielectric medium. We represent the quantum flow in terms of the non-equal time commutators involving electric and magnetic fields, as well as the local displacement field. The commutators are calculated applying a previously developed microscopic theory of quantization of radiation in dielectrics [22, 27–29]. Consequently, we derive from first principles the rate of spontaneous emission in an absorbing medium containing local field factors and other refractive contributions. The results support the previous introduction of the (virtual cavity) local field correction by Barnett *et al* [12, 13] on a phenomenological basis, as well as our previous approach [14] in which the spontaneous emission is treated as a resonance energy transfer to the far-zone atoms. A distinctive feature of the present formalism is that it deals with the spontaneous emission directly by calculating the quantum flow from the emitting atom. Therefore, the method is free from the divergences appearing in other approaches analysing the spontaneous decay in absorbing media.

The paper is organized as follows. In the next two subsections, we define a system of atoms interacting with a radiation field and separate the system into the emitting atom and the remaining atoms coupled with the radiation field. In subsection 2.3, we consider the time evolution of the electric and magnetic field operators and expand these operators in the powers of the interaction strength between the emitting atom and the surrounding radiation field. In subsection 2.4, we obtain the corresponding expansion of the Poynting vector. In section 3, we analyse the quantum flow due to an atom emitting both in the free space and in an absorbing medium, and obtain the corresponding rates of the spontaneous emission. In section 4, we summarize the findings.

2. General

2.1. A system of atoms coupled with a radiation field

Let us consider a system of atoms (or molecules) interacting with a quantized radiation field. We shall make use of a multipolar formulation of quantum electrodynamics (QED) in which interaction between the atoms is mediated by virtual photons, there being no direct inter-atomic coupling [30–35]. The Hamiltonian of the system in the electric dipole approximation of the multipolar QED reads

$$H = H_{\rm rad} + \sum_{\zeta} H_{\zeta} + \sum_{\zeta} V_{\zeta}, \tag{1}$$

where $H_{\rm rad}$ is the Hamiltonian of the quantized radiation field, H_{ζ} is the Hamiltonian for an atom ζ positioned at $\boldsymbol{r}_{\zeta}$ and the summation is carried out over all atoms ζ. The operator

$$V_{\zeta} = -\varepsilon_0^{-1}\boldsymbol{\mu}^{(\zeta)} \cdot \boldsymbol{d}^{\perp}(\boldsymbol{r}_{\zeta}) \tag{2}$$

describes coupling of the atom ζ with the radiation field, where $\boldsymbol{\mu}^{(\zeta)}$ is the corresponding electric dipole operator and $\boldsymbol{d}^{\perp}(\boldsymbol{r})$ is the operator for the transverse displacement field.

Both the radiation Hamiltonian and the displacement field operator can be expanded in terms of vacuum photon operators in a conventional way, see e.g. references [32–35]. Yet the vacuum photons do not represent normal modes of a coupled radiation–matter system.

2.2. Separation into zero-order Hamiltonian and interaction term

We shall be interested in the spontaneous emission by an atom (molecule) A surrounded by an atomic (molecular) medium. For this purpose, it is convenient to separate the full Hamiltonian (1) into a zero-order Hamiltonian H_0 and the interaction operator V:

$$H = H_0 + V. \tag{3}$$

The operator V describes coupling between the emitting atom A and the surrounding electromagnetic field:

$$V = -\varepsilon_0^{-1}\boldsymbol{\mu} \cdot \boldsymbol{d}^{\perp}(0), \tag{4}$$

where the emitter is assumed to be placed at the origin: $\boldsymbol{r}_A = 0$. Furthermore, we have omitted the superscript A in the dipole operator $\boldsymbol{\mu} \equiv \boldsymbol{\mu}^{(A)}$. It is worth noting that $\boldsymbol{d}^{\perp}(\boldsymbol{r})$ is calculated at the emitter site $\boldsymbol{r} = 0$ in equation (4). Therefore, it is the local displacement operator that is featured in the interaction operator V.

The zero-order Hamiltonian H_0 comprises the Hamiltonian for the emitter H_A and the Hamiltonian $H_{\rm pol}$ for the radiation field coupled with the remaining species $\zeta \neq A$:

$$H_0 = H_A + H_{\rm pol}, \tag{5}$$

$$H_{\rm pol} = H_{\rm rad} + \sum_{\zeta \neq A} H_{\zeta} + \sum_{\zeta \neq A} V_{\zeta}. \tag{6}$$

The normal modes of the latter radiation–matter system are known as polaritons [8, 9, 11, 22, 27–29, 36] representing photons ‘dressed’ by the atomic medium. Thus, the spontaneous decay of an atom involves emission of a polariton (a medium-dressed photon) rather than a vacuum photon.

2.3. Time evolution of the field operators

Let us consider the time evolution of the electric and magnetic field operators in the Heisenberg-interaction representation ($\boldsymbol{x}$ stands for $\boldsymbol{e}$ or $\boldsymbol{h}$):

$$\tilde{\boldsymbol{x}}(\boldsymbol{r},t) = \mathrm{e}^{-\mathrm{i}H_0 t/\hbar}\,\mathrm{e}^{\mathrm{i}Ht/\hbar}\boldsymbol{x}(\boldsymbol{r})\,\mathrm{e}^{-\mathrm{i}Ht/\hbar}\,\mathrm{e}^{\mathrm{i}H_0 t/\hbar}, \tag{7}$$

where $\boldsymbol{x}(\boldsymbol{r})$ is the corresponding Schrödinger field. The operator $\tilde{\boldsymbol{x}}(\boldsymbol{r},t)$ obeys the following equation of motion:

$$\mathrm{i}\hbar\frac{\partial\tilde{\boldsymbol{x}}(\boldsymbol{r},t)}{\partial t} = [\tilde{\boldsymbol{x}}(\boldsymbol{r},t), V(t)], \tag{8}$$

with $V(t) = \exp(-\mathrm{i}H_0 t/\hbar)V\exp(\mathrm{i}H_0 t/\hbar)$. We shall expand $\tilde{\boldsymbol{x}}(\boldsymbol{r},t)$ in the powers of the interaction strength:

$$\tilde{\boldsymbol{x}}(\boldsymbol{r},t) = \boldsymbol{x}^{(0)}(\boldsymbol{r},t) + \boldsymbol{x}^{(1)}(\boldsymbol{r},t) + \boldsymbol{x}^{(2)}(\boldsymbol{r},t) + \cdots, \tag{9}$$

where the zero-order field coincides with the Schrödinger field $\boldsymbol{x}^{(0)}(\boldsymbol{r},t) = \boldsymbol{x}(\boldsymbol{r}) \equiv \boldsymbol{x}(\boldsymbol{r},0)$. The higher order fields can be obtained through the following recurrent relationship:

$$\boldsymbol{x}^{(j+1)}(\boldsymbol{r},t) = \mathrm{i}(\hbar\varepsilon_0)^{-1}\int_0^t \mathrm{d}t'\big[\boldsymbol{x}^{(j)}(\boldsymbol{r},t'), d_l^{\perp}(0,-t')\mu^l(-t')\big] \qquad (j \geqslant 0), \tag{10}$$

where the summation over a repeated Cartesian index l is implied. Here,

$$\boldsymbol{\mu}(\tau) = \mathrm{e}^{\mathrm{i}H_A\tau/\hbar}\boldsymbol{\mu}\,\mathrm{e}^{-\mathrm{i}H_A\tau/\hbar}$$

and

$$\boldsymbol{d}^{\perp}(\boldsymbol{r}_A,\tau) = \mathrm{e}^{\mathrm{i}H_{\mathrm{pol}}\tau/\hbar}\boldsymbol{d}^{\perp}(\boldsymbol{r}_A)\,\mathrm{e}^{-\mathrm{i}H_{\mathrm{pol}}\tau/\hbar}$$

are operators in the interaction representation.

Since $\big[\boldsymbol{x}(\boldsymbol{r},0), d_l^{\perp}(0,-t')\big]$ is a c-number, equation (10) yields the following first- and second-order fields:

$$\boldsymbol{x}^{(1)}(\boldsymbol{r},t) = \mathrm{i}(\hbar\varepsilon_0)^{-1}\int_0^t \mathrm{d}t'\big[\boldsymbol{x}(\boldsymbol{r},0), d_l^{\perp}(0,-t')\big]\mu^l(-t'), \tag{11}$$

$$\boldsymbol{x}^{(2)}(\boldsymbol{r},t) = -(\hbar\varepsilon_0)^{-2}\int_0^t \mathrm{d}t'\big[\boldsymbol{x}(\boldsymbol{r},0), d_l^{\perp}(0,-t')\big]\int_0^{t'}\mathrm{d}t''d_j^{\perp}(0,-t'')[\mu^l(-t'),\mu^j(-t'')]. \tag{12}$$

2.4. Poynting vector

We are interested in a flow of the emitting energy from the source atom A. The operator of the Poynting vector in the Heisenberg-interaction representation reads

$$\tilde{\boldsymbol{S}} = \tfrac{1}{2}(\tilde{\boldsymbol{e}}\times\tilde{\boldsymbol{h}} - \tilde{\boldsymbol{h}}\times\tilde{\boldsymbol{e}}). \tag{13}$$

Using the Poynting vector, one can calculate the rate of the spontaneous emission by analysing the total flow of energy over a sphere of a radius r [25, 26]:

$$J_\gamma = \int r\langle\mathrm{vac}|\langle\gamma|\tilde{\boldsymbol{S}}|\gamma\rangle|\mathrm{vac}\rangle\cdot\boldsymbol{r}\,\mathrm{d}\Omega, \tag{14}$$

where $|\gamma\rangle|\mathrm{vac}\rangle$ is the initial state of the whole system in which the emitting atom A is in an excited state $|\gamma\rangle$ and the surrounding polariton medium in its ground (vacuum) state $|\mathrm{vac}\rangle$.

The electric and magnetic field operators entering equation (13) can be expanded using the procedure outlined in the previous subsection. As a result, one obtains the expansion of

the Poynting vector in the powers of the interaction strength between the emitting atom and the surrounding polariton bath. The second-order Poynting vector reads

$$\tilde{\boldsymbol{S}}^{(2)} = \tilde{\boldsymbol{S}}^{(1,1)} + \tilde{\boldsymbol{S}}^{(2,0)} + \tilde{\boldsymbol{S}}^{(0,2)}, \tag{15}$$

with

$$\tilde{\boldsymbol{S}}^{(q,r)} = \tfrac{1}{2}\, \boldsymbol{e}^{(q)} \times \boldsymbol{h}^{(r)} + \text{h.c.} \tag{16}$$

Here, $\boldsymbol{e}^{(q)}$ and $\boldsymbol{h}^{(r)}$ are the qth-order electric and the rth-order magnetic fields. Note that the zero- and first-order Poynting vectors $\boldsymbol{S}^{(0)} = \boldsymbol{S}^{(0,0)}$ and $\boldsymbol{S}^{(1)} = \boldsymbol{S}^{(0,1)} + \boldsymbol{S}^{(1,0)}$ do not contribute to the energy flow (14) and hence are out of interest. Therefore in what follows we shall concentrate on the second-order Poynting vector.

The quantum mechanical average of the Poynting vector can be expressed as

$$\langle \text{vac}|\langle\gamma|\tilde{\boldsymbol{S}}^{(2)}|\gamma\rangle|\text{vac}\rangle = \boldsymbol{A}^{\gamma} + \boldsymbol{B}^{\gamma} \tag{17}$$

where $\boldsymbol{A}^{\gamma} = \langle \text{vac}|\langle\gamma|\tilde{\boldsymbol{S}}^{(1,1)}|\gamma\rangle|\text{vac}\rangle$ is a contribution due to the first-order electric and magnetic fields, $\boldsymbol{B}^{\gamma} = \langle \text{vac}|\langle\gamma|(\tilde{\boldsymbol{S}}^{(2,0)} + \tilde{\boldsymbol{S}}^{(0,2)})|\gamma\rangle|\text{vac}\rangle$ being due to the products of the zero- and second-order fields.

The former $\boldsymbol{A}^{\gamma}$ reads

$$\boldsymbol{A}^{\gamma} = \frac{1}{2}\sum_{\kappa} \boldsymbol{e}^{(1)}_{\gamma\kappa} \times \boldsymbol{h}^{(1)}_{\kappa\gamma} + \text{c.c.} \tag{18}$$

where

$$\boldsymbol{e}^{(1)}_{\gamma\kappa} = \mathrm{i}(\hbar\varepsilon_0)^{-1}\mu^{l}_{\gamma\kappa}\int_0^t \mathrm{d}t'\big[\boldsymbol{e}(\boldsymbol{r},0), d_l^{\perp}(0,-t')\big]\,\mathrm{e}^{\mathrm{i}\omega_{\kappa\gamma}t'}, \tag{19}$$

$$\boldsymbol{h}^{(1)}_{\kappa\gamma} = \mathrm{i}(\hbar\varepsilon_0)^{-1}\mu^{j}_{\kappa\gamma}\int_0^t \mathrm{d}t'\big[\boldsymbol{h}(\boldsymbol{r},0), d_j^{\perp}(0,-t')\big]\,\mathrm{e}^{\mathrm{i}\omega_{\gamma\kappa}t'}, \tag{20}$$

$\omega_{\gamma\kappa} = \omega_{\gamma} - \omega_{\kappa}$ is the frequency of the atomic transition $\gamma \to \kappa$ and $\boldsymbol{\mu}_{\gamma\kappa}$ is the corresponding transition dipole moment.

For times exceeding the inverse emission frequencies $\omega_{\gamma\kappa}^{-1}$, one can neglect the fast oscillating terms in the latter $\boldsymbol{B}^{\gamma}$ giving

$$\boldsymbol{B}^{\gamma} = \frac{1}{2}\sum_{\kappa} \text{sign}\,(\omega_{\gamma\kappa})\, \boldsymbol{e}^{(1)}_{\gamma\kappa} \times \boldsymbol{h}^{(1)}_{\kappa\gamma} + \text{c.c.} \tag{21}$$

where $\text{sign}(x) = x/|x|$ is a sign function. Therefore, the full second-order Poynting vector then reads

$$\langle \text{vac}|\langle\gamma|\tilde{\boldsymbol{S}}^{(2)}|\gamma\rangle|\text{vac}\rangle = \sum_{\kappa} \Theta(\omega_{\gamma\kappa})\, \boldsymbol{e}^{(1)}_{\gamma\kappa} \times \boldsymbol{h}^{(1)}_{\kappa\gamma} + \text{c.c.} \tag{22}$$

where $\Theta(\cdots)$ is a unit step function.

It is instructive that contributions due to the upward atomic transitions cancel out in equation (22) due to the term $\boldsymbol{B}^{\gamma}$ featured in equation (17). Therefore, only downward transitions contribute to the Poynting vector (22) as required. Furthermore, the term $\boldsymbol{B}^{\gamma}$ gives the second half to the energy flow associated with the downward atomic transitions. This shows the importance of the terms $\tilde{\boldsymbol{S}}^{(2,0)}$ and $\tilde{\boldsymbol{S}}^{(0,2)}$ due to the zero- and second-order electric and magnetic fields, the point already discussed by Power and Thirunamachandran [25, 26] in their analysis of quantized fields generated by a source atom in vacuum. Note that our analysis is not restricted to the case where the source atom is situated in the free space. The influence of the material medium appears through the non-equal time commutators featured in equations (19) and (20). In the next subsection we shall outline the analysis of the quantum flow from a source atom in the free space [25, 26]. Subsequently, we shall extend the treatment to the case of an absorbing dielectric medium.

3. Spontaneous emission in free space and media

3.1. Emission in the free space

In the case of the free space, the non-equal time commutators featured in equations (19) and (20) can be easily obtained using the familiar expansion for the field operators in terms of vacuum photons [32–34] giving

$$\left[e_q(\boldsymbol{r},0), d_l^{\perp}(0,-t')\right] = \mathrm{i}\hbar c(\nabla^2\delta_{ql} - \nabla_q\nabla_l)\Delta(r,t'), \tag{23}$$

$$\left[h_s(\boldsymbol{r},0), d_j^{\perp}(0,-t')\right] = \mathrm{i}\hbar c\varepsilon_0\varepsilon_{sjp}\nabla_p\frac{\partial}{\partial t'}\Delta(r,t'), \tag{24}$$

where

$$\Delta(r,t') = \frac{\delta(r-ct') - \delta(r+ct')}{4\pi r}, \tag{25}$$

is the relativistic Δ function [37]. Hence, one has for times in excess of the relativistic time lag, $t > r/c$,

$$\left(e^{(1)}_{\gamma\kappa}\right)_q = \mu^l_{\gamma\kappa}(-\nabla^2\delta_{ql} + \nabla_q\nabla_l)\frac{\mathrm{e}^{\mathrm{i}k_{\kappa\gamma}r}}{4\pi\varepsilon_0 r} = \frac{\mu^l_{\gamma\kappa}}{4\pi\varepsilon_0}k^3_{\kappa\gamma}f_{ql}(k_{\kappa\gamma}\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}k_{\kappa\gamma}r}, \tag{26}$$

$$\left(h^{(1)}_{\kappa\gamma}\right)_s = \mathrm{i}\omega_{\gamma\kappa}\mu^j_{\kappa\gamma}\varepsilon_{sjp}\nabla_p\frac{\mathrm{e}^{\mathrm{i}k_{\gamma\kappa}r}}{4\pi r} = \omega_{\gamma\kappa}\frac{\mu^j_{\kappa\gamma}}{4\pi}k^2_{\gamma\kappa}g_{sj}(k_{\kappa\gamma}\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}k_{\gamma\kappa}r}, \tag{27}$$

where

$$f_{ql}(k_{\kappa\gamma}\boldsymbol{r}) = \left[\beta_{ql}\left(\frac{1}{k^3_{\kappa\gamma}r^3} - \frac{\mathrm{i}}{k^2_{\kappa\gamma}r^2}\right) - \alpha_{ql}\frac{1}{k_{\kappa\gamma}r}\right]\mathrm{e}^{\mathrm{i}k_{\kappa\gamma}r}, \tag{28}$$

$$g_{sj}(k_{\kappa\gamma}\boldsymbol{r}) = -\varepsilon_{sjp}\hat{r}_p\left[\frac{1}{k^2_{\gamma\kappa}r^2} - \frac{\mathrm{i}}{k_{\gamma\kappa}r}\right]\mathrm{e}^{\mathrm{i}k_{\gamma\kappa}r} \tag{29}$$

are the tensors describing the dipole radiation. Here,

$$\alpha_{ql} = \delta_{ql} - \hat{r}_q\hat{r}_l, \qquad \beta_{ql} = \delta_{ql} - 3\hat{r}_q\hat{r}_l, \tag{30}$$

where $\hat{\boldsymbol{r}} = \boldsymbol{r}/r$ is a unit vector and $k_{\gamma\kappa} = \omega_{\gamma\kappa}/c$ is a wavevector corresponding to the atomic transition $\gamma \to \kappa$.

Consequently, the Poynting vector, equation (22), takes the form

$$\langle\mathrm{vac}|\langle\gamma|\tilde{\boldsymbol{S}}^{(2)}|\gamma\rangle|\mathrm{vac}\rangle = \hat{\boldsymbol{r}}\sum_{\kappa}\omega_{\gamma\kappa}\Theta(\omega_{\gamma\kappa})\frac{k^3_{\gamma\kappa}}{8\pi^2\varepsilon_0 r^2}[|\mu_{\kappa\gamma}|^2 - (\boldsymbol{\mu}_{\kappa\gamma}\cdot\hat{\boldsymbol{r}})^2], \tag{31}$$

where the terms with r^{-4} and r^{-6} cancel exactly in the Poynting vector (31) leaving only the r^{-2} term [25, 26]. Therefore, the result (31) does not necessarily restrict r to large distances. Substituting equation (31) into (14), the quantum flow can be represented as

$$J_{\gamma} = \sum_{\kappa}\hbar\omega_{\gamma\kappa}\Gamma^{\mathrm{vac}}_{\gamma\to\kappa}\Theta(\omega_{\gamma\kappa}), \tag{32}$$

where

$$\Gamma^{\mathrm{vac}}_{\gamma\to\kappa} = \frac{\omega^3_{\gamma\kappa}|\mu_{\kappa\gamma}|^2}{3\pi c^3\hbar\varepsilon_0} \tag{33}$$

is the vacuum rate of the spontaneous emission associated with the atomic transition $\gamma \to \kappa$. Such a method of obtaining the emission rate [25, 26] can be contrasted with a conventional approach employing the Fermi golden rule [38, 39].

3.2. Emission in an absorbing medium

Consider now the case where the emitter is in a dielectric medium. The non-equal time commutators featured in equations (19) and (20) can then be obtained using a microscopically derived expansion of the electric and magnetic field operators $e_q(\boldsymbol{r}, 0)$ and $h_s(\boldsymbol{r}, 0)$, as well as the local displacement field operator $d_l^{\perp}(0, -t')$ in terms of polariton creation and annihilation operators [27, 28]. Alternatively, non-equal time commutators can be calculated adapting for the present situation a Green function technique developed in [22, 29]. In fact, the commutators can be represented in terms of the appropriate (two-site) Green functions:

$$\left[x_u(\boldsymbol{r}, 0), d_l^{\perp}(0 - t')\right] = \frac{1}{2\pi \mathrm{i}} \int_{-\infty}^{+\infty} \mathrm{d}\omega \left[G_{ul}^{xd}(\boldsymbol{r}, \omega - \mathrm{i}s) - G_{ul}^{xd}(\boldsymbol{r}, \omega + \mathrm{i}s)\right] \mathrm{e}^{\mathrm{i}\omega t'}, \tag{34}$$

where $s \to +0$,

$$G_{ul}^{xd}(\boldsymbol{r}, \omega) = \langle \mathrm{vac} | \left\{ x_u(\boldsymbol{r}) G_+(\omega) d_l^{\perp}(0) - d_l^{\perp}(0) G_-(\omega) x_u(\boldsymbol{r}) \right\} | \mathrm{vac} \rangle, \tag{35}$$

and $G_{\pm}(\omega) = (\omega \pm H_{\mathrm{pol}}/\hbar)^{-1}$. Here, x stands for the electric and magnetic field operators: $x = e, h$. For $x = d$ the Green function $G_{ul}^{xd}(\mathbf{r}, \omega)$ has been calculated in [22, 29] assuming that the host atoms are of the same type, placed regularly to form a simple cubic lattice, and characterized by isotropic polarizabilities. The analysis can be extended to the case where $x = e, h$ giving

$$\left[e_q(\boldsymbol{r}, 0), d_l^{\perp}(0, -t')\right] = \mathrm{i}\hbar c (\nabla^2 \delta_{ql} - \nabla_q \nabla_l) \tilde{\Delta}_{(e)}(r, t'), \tag{36}$$

$$\left[h_s(\boldsymbol{r}, 0), d_j^{\perp}(0, -t')\right] = \mathrm{i}\hbar c \varepsilon_0 \varepsilon_{sjp} \nabla_p \frac{\partial}{\partial t'} \tilde{\Delta}_{(h)}(r, t'), \tag{37}$$

with

$$\tilde{\Delta}_{(x)}(r, t') = \frac{\tilde{\delta}_{(x)}(r, t') - \tilde{\delta}_{(x)}^*(r, -t')}{4\pi r}, \tag{38}$$

where

$$\tilde{\delta}_{(e)}(r, t') = \frac{1}{2\pi c} \int_{-\infty}^{+\infty} \mathrm{d}\omega \left(\frac{n^2 + 2}{3}\right) \mathrm{e}^{\mathrm{i}\omega(nr/c - t')}, \tag{39}$$

$$\tilde{\delta}_{(h)}(r, t') = \frac{1}{2\pi c} \int_{-\infty}^{+\infty} \mathrm{d}\omega \left(\frac{n^2 + 2}{3n^2}\right) \mathrm{e}^{\mathrm{i}\omega(nr/c - t')} \tag{40}$$

are the smoothened delta functions and $n \equiv n(\omega) = n' + \mathrm{i}n''$ is a complex refractive index satisfying the Clausius–Mosotti relation. The refractive index $n(\omega)$ contains no singularities in the upper half plane and exhibits other general properties, such as $n(-\omega) = n^*(\omega)$ and $n'' > 0$ for $\omega > 0$. In the vacuum case ($n = 1$), the modified delta functions reduce to usual retarded and advanced delta functions: $\tilde{\delta}_{(e)}(r, \pm t') = \tilde{\delta}_{(f)}(r, \pm t') = \delta(r \pm ct')$.

The integrands in equations (39) and (40) have no singularities in the upper half plane, so $\tilde{\delta}_{(x)}(r, t')$ vanishes for $t' - r/c < 0$. In a similar manner, $\tilde{\delta}_{(x)}^*(r, -t')$ vanishes for $t' + r/c > 0$ and hence does not contribute to the integrals featured in equations (19) and (20). Using equations (36) and (37), the integrals of interest for times in excess of the time lag read

$$\int_0^t \mathrm{d}t' \left[e_q(\boldsymbol{r}, 0), d_l^{\perp}(0, -t')\right] \mathrm{e}^{\mathrm{i}\omega_{\kappa\gamma} t'} = \mathrm{i}\hbar \left(\frac{n_{\kappa\gamma}^2 + 2}{3n_{\kappa\gamma}^2}\right) (\nabla^2 \delta_{ql} - \nabla_q \nabla_l) \frac{\mathrm{e}^{\mathrm{i}k_{\kappa\gamma} r}}{4\pi r}, \tag{41}$$

$$\int_0^t \mathrm{d}t' \left[h_s(\boldsymbol{r}, 0), d_j^{\perp}(0, -t')\right] \mathrm{e}^{\mathrm{i}\omega_{\gamma\kappa} t'} = \hbar \varepsilon_0 \left(\frac{n_{\gamma\kappa}^2 + 2}{3}\right) \omega_{\gamma\kappa} \varepsilon_{sjp} \nabla_p \frac{\mathrm{e}^{\mathrm{i}k_{\kappa\gamma} r}}{4\pi r}, \tag{42}$$

where $k_{\kappa\gamma} = \omega_{\kappa\gamma} n_{\kappa\gamma}/c$ is a complex wavevector, $n_{\gamma\kappa} = n(\omega_{\gamma\kappa})$ is a refractive index calculated at the emission frequency $\omega_{\gamma\kappa}$ and $n_{\kappa\gamma} = n(\omega_{\kappa\gamma}) = n_{\gamma\kappa}^*$ is its complex conjugate

counterpart. In calculating the above integrals, we have assumed that the refractive index $n(\omega)$ is a sufficiently smooth function of frequency. Consequently, the major contribution to the integration over the frequency comes from a narrow area around $\omega = \omega_{\kappa\gamma}$ or $\omega = \omega_{\gamma\kappa}$ leading to the results (41) or (42).

Substituting equations (41) and (42) into equations (19) and (20), one finds

$$\left(e^{(1)}_{\gamma\kappa}\right)_q = \left(\frac{n^2_{\kappa\gamma}+2}{3n^2_{\kappa\gamma}}\right)\frac{\mu^l_{\gamma\kappa}}{4\pi\varepsilon_0}k^3_{\kappa\gamma}\, f_{ql}(k_{\kappa\gamma}\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}k_{\kappa\gamma}r}, \tag{43}$$

$$\left(h^{(1)}_{\kappa\gamma}\right)_s = \omega_{\gamma\kappa}\left(\frac{n^2_{\gamma\kappa}+2}{3}\right)\frac{\mu^j_{\kappa\gamma}}{4\pi}k^2_{\gamma\kappa}\, g_{sj}(k_{\kappa\gamma}\boldsymbol{r})\,\mathrm{e}^{\mathrm{i}k_{\gamma\kappa}r}. \tag{44}$$

where the tensors $f_{ql}(k_{\kappa\gamma}\boldsymbol{r})$ and $g_{sj}(k_{\kappa\gamma}\boldsymbol{r})$ have the same form as in the previously considered vacuum case, the medium effects coming from the modification of the wavevector $k_{\kappa\gamma} = \omega_{\kappa\gamma}n_{\kappa\gamma}/c$. In such a situation, the Poynting vector (22) reads, for $|k_{\gamma\kappa}|r \gg 1$,

$$\langle\mathrm{vac}|\langle\gamma|\tilde{\boldsymbol{S}}^{(2)}|\gamma\rangle|\mathrm{vac}\rangle = \frac{3\hbar\hat{\boldsymbol{r}}}{8\pi r^2}\sum_{\kappa<\gamma}\omega_{\gamma\kappa}\Gamma^{\mathrm{med}}_{\gamma\to\kappa}[|\mu_{\kappa\gamma}|^2 - (\boldsymbol{\mu}_{\kappa\gamma}\cdot\hat{\boldsymbol{r}})^2]\,\mathrm{e}^{-\alpha_{\gamma\kappa}r}, \tag{45}$$

where $\alpha_{\gamma\kappa} = 2\omega_{\gamma\kappa}n''_{\gamma\kappa}r/c$ is the Beer length absorbency at the emission frequency $\omega_{\gamma\kappa}$ and

$$\Gamma^{\mathrm{med}}_{\gamma\to\kappa} = n'_{\kappa\gamma}\left|\frac{n^2_{\gamma\kappa}+2}{3}\right|^2\frac{\omega^3_{\gamma\kappa}|\mu_{\kappa\gamma}|^2}{3\pi c^3\hbar\varepsilon_0}. \tag{46}$$

For an absorbing medium the result (45) holds for the far-zone distances only. In the near zone, additional terms r^{-n} with $n > 2$ appear due to the non-radiative losses. The non-radiative contributions become negligible in the far zone ($|k_{\gamma\kappa}|r \gg 1$) in which the Poynting vector (45) describes the radiative energy flow proportional to r^{-2} and hence can be attributed to the spontaneous emission. Substituting equation (45) into (14), the quantum flow of energy reads

$$J_\gamma = \sum_{\kappa<\gamma}\hbar\omega_{\gamma\kappa}\Gamma^{\mathrm{med}}_{\gamma\to\kappa}\,\mathrm{e}^{-\alpha_{\gamma\kappa}r}. \tag{47}$$

Thus, $\Gamma^{\mathrm{med}}_{\gamma\to\kappa}$ can be identified as the rate of the spontaneous emission is a dielectric medium.

The energy flow (47) appears to be affected by the dielectric medium both through the exponential factor proportional to $\mathrm{e}^{-\alpha_{\gamma\kappa}r}$ (describing absorption losses in the medium) and also through the refractive modifications of the emission rate $\Gamma^{\mathrm{med}}_{\gamma\to\kappa}$. The latter $\Gamma^{\mathrm{med}}_{\gamma\to\kappa}$ contains local field factors and other refractive contributions. The result (46) supports the previous introduction of the (virtual cavity) local field correction by Barnett *et al* [12, 13] on a phenomenological basis, as well as our previous approach [14] in which the spontaneous emission is considered as a far-zone resonance energy transfer [22].

4. Conclusions

We have considered the spontaneous emission in an absorbing medium by calculating the quantum flow of energy from an emitting atom. For this purpose, we have extended the analysis by Power and Thirunamachandran of quantized Maxwell fields in a vicinity of an atom in the free space [25, 26] to a situation where the emitter is in a dielectric medium. We have represented the quantum flow in terms of the non-equal time commutators involving electric and magnetic fields, as well as the local displacement field. The commutators

have been calculated applying a previously developed microscopic theory of quantization of radiation in dielectrics [22, 27–29]. Consequently, we have derived from first principles the rate of spontaneous emission in an absorbing medium containing local field factors and other refractive contributions. The results support the previous introduction of the (virtual cavity) local field correction by Barnett *et al* [12, 13] on a phenomenological basis, as well as our previous approach [14] in which the spontaneous emission is considered as a resonance energy transfer to the far-zone atoms. A distinctive feature of the present formalism is that it deals with the spontaneous emission directly by calculating the quantum flow from the emitting atom. Therefore, the method is free from the divergences appearing in other approaches analysing the spontaneous decay.

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