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Local-field effects on spontaneous emission of impurity atoms in homogeneous dielectrics

Frank J.P. Schuurmans *, Pedro de Vries [1], Ad Lagendijk

Van der Waals-Zeeman Instituut, Universiteit van Amsterdam, Valckenierstraat 65, NL-1018 XE Amsterdam, the Netherlands

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Abstract

The local-field corrections to the spontaneous emission rate of an impurity atom in an otherwise homogeneous dielectric are calculated by making use of a macroscopic approach. An extension of a method introduced by Onsager and Böttcher is presented. By distinguishing between the substitutional and interstitial character of impurities, the well-known empty-cavity and Lorentz local-field factors are obtained, respectively. For fluids, the substitutional case is predicted to occur prevalently.

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In 1917, Einstein demonstrated that spontaneous emission must occur if matter and radiation are to achieve thermal equilibrium [1]. This picture is so fundamental, that spontaneous emission rates are often believed to be an inherent property of the atom. However, the spontaneous emission rate can be modified by changing the environment, as was noticed by Purcell [2]. In the early 1970s, Drexhage carried out pioneering experiments on the modification of the luminescence decay rate of Europium complexes in front of a metallic mirror [3,4]. Later, many theoretical [5,6] and experimental [7–11] investigations have been devoted to the enhancement and inhibition of spontaneous emission of atoms in resonant cavities, in which case larger modifications are possible.

Modifications of the spontaneous emission rate can also be induced by placing the radiator inside a spatially inhomogeneous dielectric. For example, the case of emission near a dielectric interface has been studied both experimentally [3,4,12–14] and theoretically [15,16]. More recently, spontaneous emission rates were predicted to alter in photonic crystals [17–19] (see also for overviews Ref. [20]), materials in which the dielectric constant is periodically varying on length scales of the wavelength of light. As a result, light in a certain frequency range cannot propagate in the crystal because of multiple Bragg reflections. At this so-called full photonic band gap, the radiative density of states (RDOS) is zero and fluorescence of excited atoms embedded in these

* Corresponding author. Tel.: +31-20-5255663, fax: +31-20-5255788, e-mail: schuurma@phys.uva.nl
[1] Present address: KNMI, Royal Netherlands Meteorological Institute, P.O. Box 201, NL-3730 AE De Bilt, the Netherlands

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materials is expected to be inhibited at those frequencies, since then coupling to propagating modes is absent. At other frequencies, it has been shown that spontaneous emission can be either enhanced or reduced [17–19] (see also for overviews Ref. [20]). Note that the atoms in question are different from the ones constituting the dielectric; such atoms will be referred to as *impurity* atoms or, equivalently, impurity dipoles.

The spontaneous emission rate $\Gamma(\varepsilon)$ of an impurity dipole inside a homogeneous medium with dielectric constant ε (at the frequency of the impurity radiation) was predicted by Nienhuis and Alkemade [21] to follow the $\sqrt{\varepsilon}$ dependence of the RDOS, and is thus given by

$$\Gamma(\varepsilon) = \sqrt{\varepsilon}\,\Gamma_0, \tag{1}$$

where Γ_0 is radiative decay rate of the dipole in vacuum. This result was obtained by quantizing the macroscopic Maxwell's equations, while in principal the dipole couples to the microscopic vacuum fluctuations. The microscopic, or local, electric field felt by a dipole can be very different from the macroscopic field (see for example Ref. [22]), and hence large deviations from Eq. (1) may be expected.

Determining the local field is an old problem in physics, for which Lorentz has given a mean-field-like argument in terms of a 'virtual' cavity surrounding a dipole of the dielectric [22,23]. For random fluids and cubic lattices, the resulting ratio of the local to the macroscopic field is given by the Lorentz local-field factor

$$\mathscr{L}_{\mathrm{Lor}} = \frac{\varepsilon + 2}{3}, \tag{2}$$

which is the same for all dipoles that constitute the dielectric. Indeed, in experiments [24] involving only *pure* systems, i.e. when only one kind of atom or molecule is present, the observed local-field effects agree very well with this Lorentz description. However, in the case of an impurity dipole, it is not at all clear whether Eq. (2) should apply.

An alternative local-field correction that has been suggested [25,26] is the so-called empty-cavity factor [22]

$$\mathscr{L}_{\mathrm{emp}} = \frac{3\varepsilon}{2\varepsilon + 1}, \tag{3}$$

since the impurity would represent an excluded volume for the dipoles constituting the dielectric. In a description of $\Gamma(\varepsilon)$ of impurity dipoles inside dielectrics, one of the factors, $\mathscr{L}_{\mathrm{Lor}}$ or $\mathscr{L}_{\mathrm{emp}}$, should be included, giving $\Gamma(\varepsilon) = \sqrt{\varepsilon}\mathscr{L}^2_{\mathrm{Lor}}\Gamma_0$ [26–33] or $\Gamma(\varepsilon) = \sqrt{\varepsilon}\mathscr{L}^2_{\mathrm{emp}}\Gamma_0$ [25,26]. Since $\Gamma(\varepsilon)$ can be expressed in terms of an expectation value of the product of two electric field operators [26], the local-field factors in $\Gamma(\varepsilon)$appear squared. The majority of the researchers in the field [27–33] favor the Lorentz factor. On the other hand, experimental results [34–36] indicate that the empty-cavity factor [25] should be employed.

Recently, we have shown [37] that for substitutional and interstitial impurities, the empty-cavity and Lorentz-local field factor apply, respectively. This crucial distinction between substitutional and interstitial character of the impurity was not made so far. Our results were obtained from a full microscopic scattering calculation using a Green's function formalism [38].

The aim of this Letter is to rederive the results of Ref. [37] in a much simpler way. First, the local-field factors for both types of impurities are calculated employing a quasi-static macroscopic approach. This calculation is in the spirit of the method used by Onsager and Böttcher to derive the local-field for pure systems [23,39]. Their method is extended here to incorporate both substitutional and interstitial impurities. Second, the polarizability is expressed in terms of a dynamic complex quantity of which the parameters are linked to one another by invoking the optical theorem, valid for elastic light scattering. In this fashion, we obtain the appropriate local-field corrections for the spontaneous emission rate of the impurities.

We first investigate the effective static polarizability of the substitutional impurity. An impurity dipole with polarizability α_{I} is placed at the center of a spherical cavity with volume Ω in an otherwise homogeneous medium with dielectric constant ε. The local-field correction pertaining to the field in the cavity is then given by [23]

$$\mathscr{L}_{\mathrm{OB}}(\alpha_{\mathrm{I}}/\Omega) = \frac{3\varepsilon}{2\varepsilon + 1 - \dfrac{2\alpha_{\mathrm{I}}}{3\Omega}(\varepsilon - 1)}. \tag{4}$$

Outside the cavity, the total static polarizability for this system is

$$\alpha_{\mathrm{sub}} = \alpha_{\mathrm{I}} \mathscr{L}_{\mathrm{emp}} \mathscr{L}_{\mathrm{OB}}(\alpha_{\mathrm{I}}/\Omega) + (1-\varepsilon)\mathscr{L}_{\mathrm{emp}}\Omega, \tag{5}$$

where the first term is the contribution of the impurity dipole α_{I}, and the second corresponds to the polarizability of the empty spherical cavity. Note that for $\alpha_{\mathrm{I}} = 0$, the static polarizability α_{sub} is not zero, that is the polarizability of the empty cavity is still remnant. For later convenience, we re-express the first contribution in Eq. (5) as

$$\alpha_{\mathrm{sub,I}} = \alpha_{\mathrm{I}} \mathscr{L}_{\mathrm{emp}}^2 \left[1 - \frac{2\alpha_{\mathrm{I}}(\varepsilon-1)\mathscr{L}_{\mathrm{emp}}}{9\varepsilon\Omega}\right]^{-1}, \tag{6}$$

where $\mathscr{L}_{\mathrm{OB}}(\alpha_{\mathrm{I}}/\Omega)$ is explicitly written in terms of $\mathscr{L}_{\mathrm{emp}}$. For $(\alpha_{\mathrm{I}}/\Omega) \ll 1$, the response of the dielectric on α_{I}, i.e. the reaction field, may be neglected. In that case the effective static polarizability of the impurity is enhanced by $\mathscr{L}_{\mathrm{emp}}^2$.

The case of the interstitial impurity dipole is modeled by placing two dipoles in the cavity. One dipole, identical to the surrounding medium dipoles, with polarizability α is positioned at center of the cavity. A different dipole, the impurity, may in principle be placed anywhere in the cavity. The local field experienced by both dipoles in the cavity comprises of four terms: a constant field $\mathscr{L}_{\mathrm{emp}}\boldsymbol{E}_{\mathrm{ext}}$ inside the cavity due to the external field $\boldsymbol{E}_{\mathrm{ext}}$, the two reaction fields produced by both dipoles (i.e. the response of the dielectric on the presence of the dipoles), and the direct interaction between the dipoles. Dekker [40] has shown that for a dipole inside a cavity the reaction field averaged over the cavity is independent of the position of the dipole. We will use this result to simplify the calculation and consider from now on fields that are averaged over the cavity, while the directions of both dipole moments are treated as being independent of one another. The reaction field for the excentrically-positioned impurity dipole is taken equal to the constant reaction field of a dipole at the center. In addition, we also average the fields experienced by a dipole due to the other one. Since the two dipoles cannot physically be at the same position, the averaging leads to a zero contribution. By symmetry, the (averaged) direction of the induced dipoles will be in the direction of the external field. The local fields computed in this way are identical to the ones for the case where the impurity dipole is put at the center also. We finally find the local-field factor for each dipole to be $\mathscr{L}_{\mathrm{OB}}((\alpha_{\mathrm{I}}+\alpha)/\Omega)$. The corresponding total static polarizability thus reads

$$\alpha_{\mathrm{int}} = (\alpha_{\mathrm{I}}+\alpha)\mathscr{L}_{\mathrm{emp}}\mathscr{L}_{\mathrm{OB}}((\alpha_{\mathrm{I}}+\alpha)/\Omega) + (1-\varepsilon)\mathscr{L}_{\mathrm{emp}}\Omega. \tag{7}$$

For $(\alpha_{\mathrm{I}}/\Omega) \to 0$, a more detailed analysis readily shows that Eq. (7) is exact in this limit. The three different contributions (impurity dipole, medium dipole, and empty cavity) are clearly identified. Contrary to the substitutional case, the total polarizability α_{int} should be zero for $\alpha_{\mathrm{I}} = 0$, as then the medium is homogeneous; effectively there is no cavity. This is indeed realized upon Onsager's assumption [39] that the inverse volume Ω^{-1} of the cavity equals the density ρ of dipoles in the medium. Consequently, the polarizability α of the medium dipole can be eliminated from Eq. (7) by using the well-known Clausius–Mossotti or Lorentz–Lorenz expression [22,23]

$$\rho\alpha = \frac{\alpha}{\Omega} = 3\frac{\varepsilon-1}{\varepsilon+2}. \tag{8}$$

In fact, α_{int} represents the effective polarizability $\alpha_{\mathrm{int,I}}$ of the interstitial impurity dipole embedded in the dielectric, that is $\alpha_{\mathrm{int}} = \alpha_{\mathrm{int,I}}$. The static polarizability of an interstitial impurity then becomes

$$\alpha_{\mathrm{int,I}} = \alpha_{\mathrm{I}} \mathscr{L}_{\mathrm{Lor}}^2 \left[1 - \frac{2\alpha_{\mathrm{I}}(\varepsilon-1)\mathscr{L}_{\mathrm{Lor}}}{9\varepsilon\Omega}\right]^{-1}, \tag{9}$$

Note that for $\alpha_{\mathrm{I}} = 0$, indeed $\alpha_{\mathrm{int,I}} = 0$, as required for the corresponding homogeneous medium. Furthermore, the structure of Eqs. (6) and (9) is identical, only $\mathscr{L}_{\mathrm{emp}}$ in Eq. (6) is replaced by $\mathscr{L}_{\mathrm{Lor}}$ in Eq. (9).

So far, only the static polarizabilities of an substitutional and interstitial impurity were calculated. In order to capture spontaneous emission of the impurity, the polarizability α_{I} of the impurity is modeled by the polarizability of a two-level system or harmonic-oscillator, obeying the optical theorem. In fact, the optical theorem may be viewed as the microscopic analogon of Einstein's thermodynamic

arguments relating absorption, stimulated and spontaneous emission. The corresponding *dynamic* polarizability of the impurity in *vacuum* is [38]

$$\alpha_I(\omega) = \alpha_I(0)\,\frac{\omega_0^2}{\omega_0^2 - \omega^2 - i\left(\Gamma_0\,\omega^3/\omega_0^2\right)}, \qquad (10)$$

Here, $\alpha_I(0)$ is the static polarizability, ω_0 the resonance frequency, and Γ_0 the resonance width, which is identical to the spontaneous emission rate. The set $\{\alpha_I(0), \omega_0, \Gamma_0\}$ has only two independent parameters, as the optical theorem relates them via [38]

$$\Gamma_0 = \alpha_I(0)\,\omega_0^4/6\pi c_0^3, \qquad (11)$$

where c_0 is the speed of light in vacuum. This relation is consistent with Fermi's Golden Rule result for Γ_0 by taking the static polarizability $\alpha_I(0) = 2|\boldsymbol{D}|^2/\varepsilon_0\hbar\omega_0$ in terms of the dipole matrix element $\boldsymbol{D}$ of two atomic levels. These relations hold for a dipole in vacuum. When considering a dipole in a background medium with dielectric constant ε and neglecting local-field or cavity effects, Eq. (11) is modified to $\Gamma(\varepsilon) = \sqrt{\varepsilon}\,\Gamma_0$, with Γ_0 determined by the polarizability and the resonance frequency as in Eq. (11). This is in agreement with the Nienhuis and Alkemade result, see Eq. (1).

Implementing the dynamic polarizability $\alpha_I(\omega)$ for the static one α_I in Eq. (6) readily gives the effective dynamic polarizability $\alpha_{\mathrm{sub,I}}(\omega)$ of a substitutional impurity

$$\alpha_{\mathrm{sub,I}}(\omega) = \alpha_I(\omega)\,\mathscr{L}_{\mathrm{emp}}^2\left[1 - \frac{2\,\alpha_I(\omega)(\varepsilon-1)\,\mathscr{L}_{\mathrm{emp}}}{9\varepsilon\Omega}\right]^{-1}. \qquad (12)$$

The polarizability of this 'dressed' impurity near resonance has a Lorentzian structure analogous to Eq. (10):

$$\alpha_{\mathrm{sub,I}}(\omega) = \alpha_{\mathrm{sub,I}}(0) \times \frac{\omega_0^2(\varepsilon)}{\omega_0^2(\varepsilon) - \omega^2 - i\left(\Gamma(\varepsilon)\,\omega^3/\omega_0^2(\varepsilon)\right)}, \qquad (13)$$

with parameters given by

$$\frac{\omega_0^2(\varepsilon)}{\omega_0^2} = \left[1 - \frac{2\,\alpha_I(0)(\varepsilon-1)\,\mathscr{L}_{\mathrm{emp}}}{9\varepsilon\Omega}\right], \qquad (14)$$

$$\alpha_{\mathrm{sub,I}}(0) = \mathscr{L}_{\mathrm{emp}}^2\,\frac{\omega_0^2}{\omega_0^2(\varepsilon)}\,\alpha_I(0), \qquad (15)$$

$$\frac{\Gamma(\varepsilon)}{\Gamma_0} = \sqrt{\varepsilon}\,\mathscr{L}_{\mathrm{emp}}^2\,\frac{\omega_0^2(\varepsilon)}{\omega_0^2}. \qquad (16)$$

Here, the spontaneous emission rate $\Gamma(\varepsilon)$ of the substitutional impurity follows from the polarizability $\alpha_{\mathrm{sub,I}}(0)$ and the resonance frequency $\omega_0(\varepsilon)$ by use of the optical theorem, see Eq. (11). It should be noted that a direct calculation of $\Gamma(\varepsilon)$ from Eq. (13) is not feasible, as the macroscopic derivation presented here is in essence quasi-static, and hence only accounts for a real polarizability. This drawback is not encountered in the full, but more complicated calculation of Ref. [37]. In vacuum, $\varepsilon = 1$, the original results are regained, that is $\omega_0(1) = \omega_0$, $\alpha_{\mathrm{sub,I}}(0) = \alpha_I(0)$, and $\Gamma(1) = \Gamma_0$. The resonance frequency is red-shifted and the line width behaves in accordance with the empty-cavity description, identical to our earlier results [37].

The dynamical properties of an interstitial impurity are obtained in a similar fashion as for the substitutional one. As a result, the polarizability of the 'dressed' interstitial impurity also has a Lorentzian line shape given by Eq. (13). The parameters for the interstitial case are

$$\frac{\omega_0^2(\varepsilon)}{\omega_0^2} = \left[1 - \frac{2\,\alpha_I(0)(\varepsilon-1)\,\mathscr{L}_{\mathrm{Lor}}}{9\varepsilon\Omega}\right], \qquad (17)$$

$$\alpha_{\mathrm{int,I}}(0) = \mathscr{L}_{\mathrm{Lor}}^2\,\frac{\omega_0^2}{\omega_0^2(\varepsilon)}\,\alpha_I(0), \qquad (18)$$

$$\frac{\Gamma(\varepsilon)}{\Gamma_0} = \sqrt{\varepsilon}\,\mathscr{L}_{\mathrm{Lor}}^2\,\frac{\omega_0^2(\varepsilon)}{\omega_0^2}. \qquad (19)$$

Again the resonance frequency is red-shifted, although with a different functional dependence on ε. The Lorentz local-field factor is found to determine the modification of the spontaneous emission rate, in agreement with Ref. [37].

The obtained results for both the substitutional and interstitial impurity still depend on the volume of the cavity Ω. For the case of the interstitials the volume is fixed: $\Omega = \rho^{-1}$ (Onsager's assumption). For substitutionals, Ω is generally taken to be the real volume of the impurity atom or molecule. For small impurity volumes, $\Omega < \rho^{-1}$, only one dipole of the dielectric is expelled from the cavity. If the impurity is large, $\Omega \gg \rho^{-1}$, many dipoles of the dielectric are removed, and the impurity is a true macroscopic cavity.

The resonance frequency of the impurity slightly shifts to the red, see Fig. 1. This shift is a direct consequence of the reaction field produced by the polarization in the medium that is induced by the impurity dipole. For small impurity polarization densities ($\alpha_I(0)/\Omega \ll 1$), the reaction field disappears, and thus the resonance frequency shift can be neglected ($\omega_0^2(\varepsilon) = \omega_0$). In experiments, this is nearly always the case, except for extremely polarizable atoms like Na, Li, Rb, so that a shift of the resonance frequency is difficult to observe.

The spontaneous emission rate increases upon placing the impurity dipole in a dielectric, see Fig. 2. The volume Ω of the cavity is irrelevant, if the impurity polarization density is small ($\alpha_I(0)/\Omega \ll 1$).

Two different classes of common host media can be distinguished: crystalline and disordered materials. In crystalline materials, which are necessarily solid, both substitutional and interstitial impurities can occur. The substitutionals are positioned at lattice sites, the interstitials somewhere in between. In the case of disordered materials, like fluids and glasses, this distinction is not so clear. For such host media, it is essential to consider the local environment in order to distinguish between substitutional and interstitial [37]. Interstitial behavior occurs when impurities do not influence the positions of the particles that form the dielectric. So the volume of the impurity has to be much smaller than the voids in between the medium dipoles. On the other hand, if the size of the impurity is comparable, or larger, than the size of the medium particles, the substitutional result applies. As, in that case, the impurity forms a real excluded volume for the dipoles of the dielectric. At very low densities, the distinction between the two types of impurities is not relevant, since any distinct feature of positional correlation of the medium dipoles will disappear, and $\mathscr{L}_{\mathrm{Lor}} \approx \mathscr{L}_{\mathrm{emp}}$ to first order in Ω^{-1}. In the case of fluids, only substitutional behavior is expected to occur, as the sizeable dimensions of the impurities present a real excluded volume for the surrounding dielectric.

In conclusion, we have investigated spontaneous emission of substitutional and interstitial impurities in dielectrics, using the Onsager–Böttcher approach.

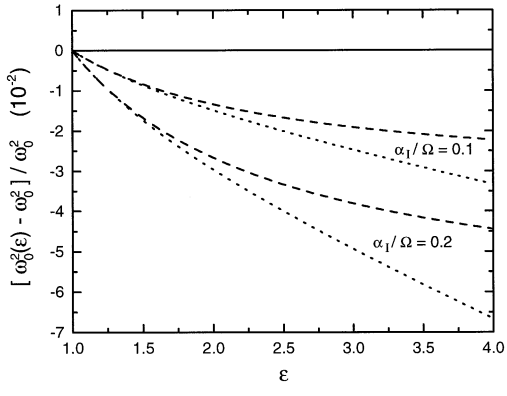

Fig. 1. The red-shift of the resonance frequency of substitutional [dashed lines, Eq. (14)] and interstitial [dotted lines, Eq. (17)] impurities, for two different polarization densities α_I/Ω. The solid line represents no shift, corresponding to $\alpha_I/\Omega \downarrow 0$. Note that the shift is rather small, only a few percent.

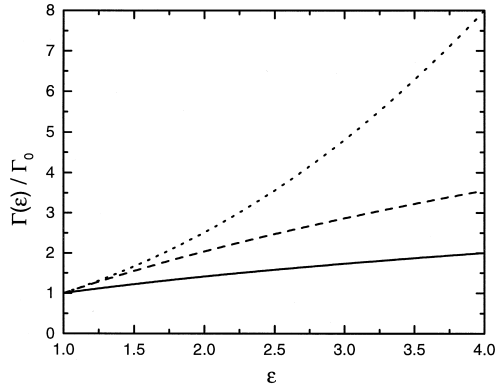

Fig. 2. The relative spontaneous emission rate of substitutional [dashed line, Eq. (16)] and interstitial [dotted line, Eq. (19)] impurities for zero resonance frequency shift, that is $\omega_0^2(\varepsilon)/\omega_0^2 = 1$. The solid line comes from Eq. (1). Large deviations from the vacuum spontaneous emission rate Γ_0 are predicted to occur in dielectrics.

The substitutional case yields the empty-cavity local-field corrections for the spontaneous emission rate, whereas for interstitials the Lorentz local-field applies.

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