Spontaneous Emission in Absorbing Dielectric Media

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(Received 27 January 1992)

We have calculated the effect of material absorption on the rate of spontaneous emission by an embedded atom using a macroscopic Green-function approach and a microscopic Hamiltonian method that includes reservoir damping. When local field effects are neglected, the free-space emission rate is modified by the *real part* of the refractive index at the transition frequency of the embedded atom. Local field effects are introduced via a local field correction factor. This modification of the spontaneous emission rate generalizes a well-known result for transparent media and is of crucial importance in assessing the degree of inhibition of spontaneous emission for frequencies close to the resonances of the dielectric.

PACS numbers: 42.50.−p, 32.70.Fw, 78.90.+t

It has long been recognized that the spontaneous emission rate of an excited atom is not an immutable property, but that it can be modified by the atomic environment [1]. The presence of suitable boundaries can enhance or reduce the local vacuum field fluctuations and thereby alter the spontaneous emission rate [2,3]. The decay rate of an excited species embedded in a bulk dielectric medium is also modified when compared with the free-space rate, given by

$$\Gamma_0 = \mu^2 \omega_a^3 / 3\pi\hbar\epsilon_0 c^3 , \tag{1}$$

for a transition of frequency ω_a and dipole moment μ. Previous analyses of the spontaneous emission rate in a bulk dielectric have treated the effect of the medium via a *real* permittivity introduced either phenomenologically [4,5] or derived from a microscopic model [6]. In the absence of absorption, the coupling between the material polarization and the radiation field introduces a band gap [7] which totally suppresses spontaneous emission [8]. However, the Kramers-Kronig relations require the permittivity in a dispersive dielectric to be complex and the region of sharpest refractive index variation usually has an associated absorption. This suggests that the absorption will play a vital role in the rate of spontaneous emission in the region corresponding to this band gap and that its effects must be included in any realistic calculation. The inclusion of dispersion and absorption should also be of relevance to the calculation of the photonic band gap which has been demonstrated in periodic dielectric structures [8,9].

In this Letter, we calculate the spontaneous emission rate in an absorbing linear dielectric by two different methods. The first is based on an application of the fluctuation-dissipation theorem to the dielectric Green function. The second proceeds from the diagonalization of a microscopic model including the electromagnetic field, the polarization of the medium, and the reservoirs that cause the polarization to decay.

The standard expression for the spontaneous emission rate for an atom at position $\mathbf{r}$ with a transition dipole matrix element μ parallel to Cartesian axis j is [10]

$$\Gamma_j = \left(\frac{2\pi}{\hbar^2}\right)\mu^2 \sum_j |\langle f|E_j^T(\mathbf{r})|0\rangle|^2 \delta(\omega-\omega_a) , \tag{2}$$

where $\mathbf{E}^T(\mathbf{r})$ is the *transverse* electric field in the Schrödinger representation and the electric field matrix element describes the accompanying excitation of the electromagnetic field from its vacuum state to a final state with a single photon of frequency ω. As this matrix element is nonzero only for single-photon states, the sum can be formally extended to all possible final states and use of the completeness of the photon states leads to

$$\Gamma_j = (2\pi/\hbar^2)\mu^2 \langle 0|E_j^T(\mathbf{r})^2|0\rangle \delta(\omega-\omega_a) . \tag{3}$$

We note that this expression is based on first-order perturbation theory (Fermi golden rule) and therefore does not include the reaction of the polarization of the medium on the atom. This approximation is equivalent to the identification of the macroscopic field $\mathbf{E}^T(\mathbf{r})$ and the local field at the position of the atom. Local field effects will be introduced at a later stage.

The field matrix element in (3) can also be expressed in the Heisenberg representation, which enables us to write the spontaneous emission rate in terms of the power spectrum of the fluctuations by

$$\Gamma_j = (2\pi/\hbar^2)\mu^2 \langle E_j^T(\mathbf{r})^2\rangle_{\omega_a} , \tag{4}$$

where $\langle E_i^T(\mathbf{r}) E_j^T(\mathbf{r}')\rangle_\omega$ is defined as the Fourier transform of the field correlation function [11]. It is readily obtained by means of the fluctuation-dissipation theorem

and Kubo's formula [11] in the form

$$\langle E_i^T(\mathbf{r})E_j^T(\mathbf{r}')\rangle_\omega = -\pi^{-1}\,\mathrm{Im}[G_{ij}^R(\mathbf{r},\mathbf{r}';\omega)]\,, \tag{5}$$

where $G_{ij}^R(\mathbf{r},\mathbf{r}';\omega)$ is the Fourier transform of the retarded Green function of the *transverse* electric field. In a homogeneous dielectric, it depends only on the difference $\mathbf{r}-\mathbf{r}'$ and is most easily calculated in reciprocal space [11,12]:

$$G_{ij}^R(\mathbf{q},\omega) = -\frac{1}{\epsilon_0}\left\{\frac{e_i(\mathbf{q},1)e_j(\mathbf{q},1)+e_i(\mathbf{q},2)e_j(\mathbf{q},2)}{(qc/\omega)^2-\epsilon(\omega)}\right\}. \tag{6}$$

Here, the $\mathbf{e}(\mathbf{q},\lambda)$ with $\lambda=1,2$ are unit independent transverse polarization vectors and $\epsilon(\omega)$ is the complex dielectric function. Substitution of the spatial Fourier transform of (6) and of (5) into (4) provides an expression for the spontaneous emission rate in which the evaluation of the angular integral gives

$$\Gamma_j = \Gamma_0 \frac{2\omega_a c^3 \epsilon''(\omega_a)}{\pi}\int_0^\infty \frac{q^2\,dq}{[\omega_a^2\epsilon'(\omega_a)-q^2c^2]^2+[\omega_a^2\epsilon''(\omega_a)]^2}\,, \tag{7}$$

where ϵ' and ϵ'' are the real and imaginary parts of the dielectric function $\epsilon(\omega)$. Note that the decay rate is independent of dipole orientation j, as expected in an isotropic medium. The remaining integral can be evaluated by contour integration. It is convenient to define the real material refractive index $\eta(\omega)$ and extinction coefficient $\kappa(\omega)$ in the usual way by

$$[\eta(\omega)+i\kappa(\omega)]^2 = \epsilon'(\omega)+i\epsilon''(\omega)\,, \tag{8}$$

and $\eta(\omega)\geq 0$. The result then takes the simple form

$$\Gamma_j = \eta(\omega_a)\Gamma_0\,. \tag{9}$$

From the above derivation, we see that the dielectric affects the spontaneous emission rate via a modification of the power spectrum of the macroscopic transverse electric field in the vacuum. This power spectrum may be calculated via the fluctuation-dissipation theorem as above or by an *ab initio* derivation from a microscopic Hamiltonian including the medium polarization and the electromagnetic field [13]. This second method relies on the explicit diagonalization of the Hamiltonian and is therefore restricted to a simple model of the dielectric. However, it can be easily extended to treat various interactions between embedded atoms or molecules.

Our starting point is the Hamiltonian density for a damped harmonic polarization coupled to the free electromagnetic field. It can be decomposed into a longitudinal part, including the longitudinal part of the electromagnetic field coupled to the longitudinal part of the polarization field, and a transverse part, including the transverse parts of both fields. As we are interested in the transverse electromagnetic field, which is not coupled to the longitudinal part, we consider only the transverse part of the Hamiltonian [for notational simplicity, we do not write the $(\mathbf{r},t)$ dependence of all the operators explicitly]:

$$\hat{\mathcal{H}} = \frac{1}{2}\left\{\epsilon_0(\hat{\mathbf{E}}^T)^2+\frac{1}{\mu_0}(\nabla\times\hat{\mathbf{A}})^2+\frac{\alpha^2}{\rho}\hat{\mathbf{A}}^2\right\}+\sum_i\int d\omega\,\hbar\omega\hat{B}_i^\dagger(\omega)\hat{B}_i(\omega)$$
$$+\left(\frac{\hbar\alpha^2}{2\rho}\right)^{1/2}\sum_i\hat{A}_i\int d\omega[\zeta^*(\omega)\hat{B}_i(\omega)+\zeta(\omega)\hat{B}_i^\dagger(\omega)]\,, \tag{10}$$

where α is the charge density, ρ is the effective mass density associated with the harmonic polarization, and $\hat{\mathbf{A}}$ is the vector potential. The $\hat{B}_i(\omega)$ are annihilation operators for the i component of the transverse polarization field, dressed by its interaction with the reservoirs responsible for the damping of the polarization, and $\zeta(\omega)$ is the coupling between the field and the dressed polarization. These operators satisfy the usual equal-time commutation relations for transverse fields (we now write the space-time dependence explicitly):

$$[\hat{B}_i(\mathbf{r},t,\omega),\hat{B}_j^\dagger(\mathbf{r}',t,\omega')] = \delta_{ij}^T(\mathbf{r}-\mathbf{r}')\delta(\omega-\omega')\,. \tag{11}$$

The fields in the Hamiltonian density can be expressed as Fourier expansions in single-mode creation and annihilation operators in the normal way. The Hamiltonian is then determined by integrating the density over all space. It can be diagonalized exactly by application of a technique developed by Fano [14]. This procedure involves seeking annihilation operators of the form $\hat{C}_\lambda(\mathbf{q},\omega)$ expressed as linear combinations of the field and matter creation and annihilation operators and obeying the commutation relations

$$[\hat{C}_\lambda(\mathbf{q},\omega),\hat{C}_{\lambda'}^\dagger(\mathbf{q}',\omega')] = \delta_{\lambda,\lambda'}\delta(\mathbf{q}-\mathbf{q}')\delta(\omega-\omega')\,, \tag{12}$$

such that the Hamiltonian has the form

$$\hat{H} = \int d^3q\int d\omega\sum_\lambda \hbar\omega\hat{C}_\lambda^\dagger(\mathbf{q},\omega)\hat{C}_\lambda(\mathbf{q},\omega)\,. \tag{13}$$

These annihilation and creation operators have simple

harmonic dependence and they act to annihilate or create dressed excitations of the interacting medium and the electromagnetic field. The physical properties of the medium and the electromagnetic field can be expressed in terms of the dressed operators; in particular, we find that the transverse electric field has the form

$$\hat{\mathbf{E}}^T(\mathbf{r},t)=\frac{1}{(2\pi)^{3/2}}\int d^3q\sum_\lambda \mathbf{e}(\mathbf{q},\lambda)\left(\frac{\hbar a^2}{2\rho\epsilon_0^2}\right)^{1/2}\int d\omega\left[-\frac{i\omega\zeta^*(\omega)}{\omega^2\epsilon(\omega)-q^2c^2}\hat{C}_\lambda(\mathbf{q},\omega)e^{-i(\omega t-\mathbf{q}\cdot\mathbf{r})}+\text{H.c.}\right], \tag{14}$$

where the complex dielectric constant $\epsilon(\omega)$ is related to the microscopic properties of the model by the relation

$$\epsilon(\omega)=1+\frac{a^2}{2\rho\epsilon_0\omega^2}\left[\mathcal{P}\int_{-\infty}^{\infty}\frac{\omega|\zeta(\omega')|^2}{\omega'(\omega'-\omega)}d\omega'+i\pi|\zeta(\omega)|^2\right], \tag{15}$$

and it can be easily shown that $\epsilon(\omega)$ satisfies the Kramers-Kronig relations [13].

Using the expression for the transverse electric field in (14), we substitute in (3) to calculate the fluctuations in the ground state of the coupled matter-field system (the dielectric vacuum) and regain the result that the spontaneous emission rate in the dielectric differs from the free-space rate by a factor equal to the real part of the refractive index.

As mentioned earlier, the above calculations are incomplete since they determine the spontaneous emission rate from the power spectrum of the fluctuations in the *macroscopic* field $\mathbf{E}^T(\mathbf{r})$ rather than the *local* field at the position of the atom. The relation between the local and macroscopic fields depends upon the precise nature of the environment of the radiating atom, and several cases have been treated for lossless dielectrics [3,5,6]. For the simple example of a virtual cavity of dimensions small compared to the length scale of the field fluctuations in a lossy dielectric, the appropriate squared local field is obtained [15] from the squared macroscopic field upon multiplication by the local field correction factor: $|[\epsilon(\omega)+2]/3|^2$. A more detailed analysis of these effects requires taking into consideration the reaction of the polarization of the medium on the emitting atom and this will be addressed in a later publication.

In Fig. 1 we plot the spontaneous emission rate of an atom embedded in a dielectric with a single resonance at ω_0 and a Lorentzian line shape as a function of the transition frequency of the atom ω_a for three different values of the loss in the dielectric. With the above local field correction factor included, the modified spontaneous emission rate is given by

$$\Gamma_j=\eta(\omega_a)|[\epsilon(\omega_a)+2]/3|^2\Gamma_0. \tag{16}$$

In the lossless case (solid curve in the figure), the dielectric has a forbidden gap between the resonance frequency ω_0 and the longitudinal frequency ω_L defined by $\omega_L^2=\omega_0^2+\omega_p^2$, ω_p being the plasma frequency of the dielectric. We note that there is a sharp rise in the decay rate for an atomic transition frequency below the dielectric resonance frequency followed by a dip above it, covering the frequency range from just above ω_0 to just above ω_L. The nonzero decay rate in the region of the band gap is only possible because of the dispersive refractive index associated with the absorption, and the inhibition of the spontaneous emission is more pronounced for the smaller absorption. As the inset of the figure shows, the regions of strong inhibition and high absorption tend to coincide, thus impeding observation of the effect. However, particularly for lower values of the loss, there is a significant range of frequencies close to ω_L for which strong inhibition is accompanied by small absorption. Another interesting possibility would be to use the enhancement of the index of refraction via quantum coherence recently

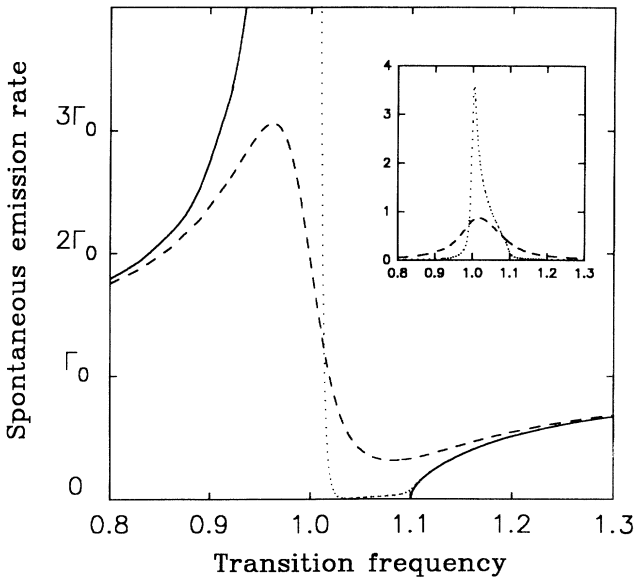

FIG. 1. Spontaneous emission rate of an embedded atom as a function of the atomic transition frequency (ω_a) for a dielectric with one resonance. Γ_0 is the free-space spontaneous emission rate and all the frequencies are in units of the resonance frequency of the dielectric ($\omega_0=1$). The dielectric constant is $\epsilon(\omega)=1+\omega_p^2/(\omega_0^2-\omega^2-i\gamma\omega)$, where ω_p is the plasma frequency chosen to give a longitudinal frequency $\omega_L=1.1$ ($\omega_p=0.46$) and γ is the loss coefficient. The three curves correspond to three values of γ: solid curve, $\gamma=0$ (no losses); dotted curve $\gamma=0.01$ (low losses); dashed curve, $\gamma=0.1$ (high losses). For frequencies below resonance, the solid curve is superimposed on the dotted one. Inset: The corresponding extinction coefficient in the dielectric.

suggested by Scully [16] to obtain a refractive index with a small real and imaginary part.

The frequency-dependent enhancement or suppression of the spontaneous emission rate can be understood as an interference between decay directly into the electromagnetic field and decay via the medium polarization. Indeed the phenomenon is reminiscent of the photoionization of an atom in which an unoccupied high-lying state is coupled to the free-electron continuum by a laser. The embedding of this bound state into the continuum causes frequency-dependent enhancement or inhibition of the ionization rate due to interference between direct ionization and ionization via the coupled bound state [17].

In conclusion, we have shown how the spontaneous emission rate of an embedded atom is modified by the dielectric. By allowing for both dispersion and absorption by the medium for the first time, our analysis enables us to cover the full frequency spectrum, including regions close to the resonances of the medium. The two methods presented here to calculate the spontaneous emission rate are in some sense complementary. The first one, based on the macroscopic Green function, does not rely on a particular model for the dielectric and can therefore be applied to any homogeneous dielectric. Its limitation is that it can only be used to calculate field expectation values which are related to the dielectric Green function. By contrast, the second method relies on the explicit introduction of a harmonic field to model the dielectric structure. However, as the expression for the electric field in the dielectric is derived, this method could be used to treat a wider range of phenomena for embedded atoms or molecules.

We thank B. J. Dalton for his helpful comments. This work was supported in part by the Royal Society of Edinburgh and by the United Kingdom Science and Engineering Research Council.

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