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SPONTANEOUS DECAY IN THE PRESENCE OF ABSORBING DIELECTRIC BODIES*

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We present a formalism for studying the influence of dispersive and absorbing dielectric bodies on a radiating atom in the framework of quantization of the phenomenological Maxwell equations for given complex permittivities of the bodies. In Markov approximation, the rate of spontaneous decay and the line shift associated with it can then be related to the complex permittivities and geometries of the bodies via the dyadic Green function of the classical boundary value problem of electrodynamics – a result which is in agreement with second-order calculations for microscopic model systems. The theory is applied to an atom near a planar interface as well as to an atom in a spherical cavity. The latter, also known as the real-cavity model for spontaneous decay of an excited atom embedded in a dielectric, is compared with the virtual-cavity model. Connections with other approaches are mentioned and the results are compared.

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1 Introduction

It is well known that the quantum statistics of photons and atoms can essentially be influenced by the presence of macroscopic bodies. In principle, such bodies can be included in the quantum mechanical description of the system as a part of matter to which the radiation field is coupled and treated microscopically, using, e.g., a canonical Hamiltonian formalism and perturbation theory (mostly up to second order). Apart from the fact that in practice the involved calculations can only be performed for relatively simple systems, each system requires its own calculation in general, because of the specific microscopic properties that must be taken into account.

In classical optics the effect of macroscopic bodies is commonly described phenomenologically in terms of a spatially varying permittivity $\epsilon(\mathbf{r},\omega)$ [or refractive index $n(\mathbf{r},\omega)=\sqrt{\epsilon(\mathbf{r},\omega)}$] which is a complex function of frequency in general. Recently a scheme for transferring this concept to quantum theory has been developed [1,2,3] which is consistent with standard QED

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as well as with the dissipation-fluctuation theorem. The advantage of such an approach is that it is valid for arbitrary linear dielectrics. Knowledge is needed only about the dispersion-absorption profile of the medium expressed in terms of the (measurable) permittivity, from which the dyadic Green function of the classical problem can be calculated. The electric and magnetic fields can then be expressed in terms of the classical Green function and a continuum of fundamental bosonic fields.

In the present paper we use that quantization scheme for studying the problem of a radiating atom in the presence of dispersive and absorbing dielectric matter. In particular, we consider the effect on the spontaneous decay of an excited atom and calculate the decay rate and the line shift. With regard to potential applications in scanning near-field optical microscopy, we present results for an atom near a planar interface. Further, we derive a correct generalization for absorbing media of the real-cavity model and the virtual-cavity model of spontaneous decay in dielectrics.

The paper is organized as follows. In Section 2 the quantization scheme for the electromagnetic field in linear Kramers-Kronig dielectrics is briefly reviewed, and general formulas for the decay rate and the line shift of an atom in the vicinity of a dielectric body are derived. In Section 3 a planar interface is considered, and Section 4 is devoted to a spherical cavity, which may serve as a model for spontaneous decay in media. The corresponding virtual-cavity model is considered in Section 5, and a comparison with the real-cavity model is given. Finally, a summary is given in Section 6.

2 Basic equations

Our analysis is based on the phenomenological quantization of the electromagnetic field in linear Kramers-Kronig dielectrics as developed in [1,2,3]. Without external sources, Maxwell's equations in (temporal) Fourier space read

$$\nabla \cdot \underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = 0, \tag{1}$$

$$\nabla \cdot \left[\epsilon_0 \epsilon(\mathbf{r},\omega) \underline{\hat{\mathbf{E}}}(\mathbf{r},\omega)\right] = \underline{\hat{\rho}}(\mathbf{r},\omega), \tag{2}$$

$$\nabla \times \underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) = i\omega \nabla \times \underline{\hat{\mathbf{B}}}(\mathbf{r},\omega), \tag{3}$$

$$\nabla \times \underline{\hat{\mathbf{B}}}(\mathbf{r},\omega) = -i\frac{\omega}{c^2}\epsilon(\mathbf{r},\omega)\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) + \mu_0 \underline{\hat{\mathbf{j}}}(\mathbf{r},\omega), \tag{4}$$

with $\epsilon(\mathbf{r},\omega) = \epsilon_R(\mathbf{r},\omega) + i\epsilon_I(\mathbf{r},\omega)$ being the spatially varying, complex permittivity of the medium which satisfies the Kramers-Kronig relations. In Eqs. (2) and (4), $\underline{\hat{\rho}}(\mathbf{r},\omega)$ and $\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega)$, respectively, are the operator noise charge and current density operators that are associated with absorption according to the dissipation-fluctuation theorem. They are related to the noise polarization $\underline{\hat{\mathbf{P}}}^N(\mathbf{r},\omega)$ as

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = -i\omega \underline{\hat{\mathbf{P}}}^N(\mathbf{r},\omega), \tag{5}$$

$$\underline{\hat{\rho}}(\mathbf{r},\omega) = -\nabla \cdot \underline{\hat{\mathbf{P}}}^N(\mathbf{r},\omega), \tag{6}$$

and hence satisfy the equation of continuity. By introducing an infinite set of bosonic field operators $\hat{\mathbf{f}}(\mathbf{r},\omega)$, all electromagnetic field operators can be expressed in terms of them using the fundamental relation

$$\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = \omega\sqrt{\frac{\hbar\epsilon_0}{\pi}\epsilon_I(\mathbf{r},\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega). \tag{7}$$

In particular, Maxwell's equations imply that the electric field operator may be written in terms of the classical dyadic Green function as

$$\underline{\hat{E}}_k(\mathbf{r},\omega) = i\mu_0\int d^3\mathbf{s}\,\omega G_{kk'}(\mathbf{r},\mathbf{r}',\omega)\underline{\hat{j}}_{k'}(\mathbf{r}',\omega), \tag{8}$$

where $G_{kk'}(\mathbf{r},\mathbf{s},\omega)$ satisfies the partial differential equation

$$\left[\partial_i^r\partial_k^r - \delta_{ik}\left(\triangle^r + \frac{\omega^2}{c^2}\,\epsilon(\mathbf{r},\omega)\right)\right] G_{kl}(\mathbf{r},\mathbf{r}',\omega) = \delta_{il}\delta(\mathbf{r}-\mathbf{r}'). \tag{9}$$

Note that the relation

$$\int d^3\mathbf{s}\,\frac{\omega^2}{c^2}\,\epsilon_I(\mathbf{s},\omega)G_{kl}(\mathbf{r},\mathbf{s},\omega)G^*_{k'l}(\mathbf{r}',\mathbf{s},\omega) = \mathrm{Im}\,G_{kk'}(\mathbf{r},\mathbf{r}',\omega) \tag{10}$$

is valid, which can be checked by post-multiplying Eq. (9) by $G^*_{ij}(\mathbf{r},\mathbf{r}'',\omega)$, adding the complex conjugate to itself and integrating over $\mathbf{r}$.

Integrating Eq. (8) with respect to ω leads to the operator of the electric field and, via Maxwell's equations, to the magnetic field operator, with the equal-time commutation relations

$$\epsilon_0\left[\hat{E}_k(\mathbf{r}),\hat{B}_l(\mathbf{r}')\right] = -i\hbar\epsilon_{klm}\partial_m^r\delta(\mathbf{r}-\mathbf{r}'), \tag{11}$$

$$\left[\hat{E}_k(\mathbf{r}),\hat{E}_l(\mathbf{r}')\right] = \left[\hat{B}_k(\mathbf{r}),\hat{B}_l(\mathbf{r}')\right] = 0, \tag{12}$$

which has been shown [3] to be valid for arbitrary linear dielectrics that are consistent with the Kramers-Kronig relations.

If there is an additional (two-level) atom present, then the total Hamiltonian of the system in dipole and rotating wave approximation can be given by

$$\hat{H} = \int d^3\mathbf{r}\int_0^\infty d\omega\,\hbar\omega\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega) + \sum_{\alpha=1}^{2}\hbar\omega_\alpha\hat{A}_{\alpha\alpha} - \left[i\omega_{21}\hat{A}_{21}\,\hat{\mathbf{A}}^{(+)}(\mathbf{r}_A)\cdot\mathbf{d}_{21} + \mathrm{H.c.}\right], \tag{13}$$

where we have chosen the Weyl gauge for the vector potential, and hence the scalar potential is set equal to zero. It can then be shown that in Markov approximation the atomic (flip) operators $\hat{A}_{\alpha\alpha'} = |\alpha\rangle\langle\alpha'|$ satisfy the following Langevin-type differential equations [4]:

$$\dot{\hat{A}}_{22} = -\Gamma\hat{A}_{22} - \left[\hat{A}_{21}\frac{\omega_{21}}{\hbar}\hat{\mathbf{A}}^{(+)}_{\mathrm{free}}(\mathbf{r}_A,t)\cdot\mathbf{d}_{21} + \mathrm{H.c.}\right] \tag{14}$$

$$\dot{\hat{A}}_{11} = -\dot{\hat{A}}_{22} \tag{15}$$

$$\dot{\hat{A}}_{21} = \left[i(\omega_{21}-\delta\omega) - \tfrac{1}{2}\Gamma\right]\hat{A}_{21} + \frac{\omega_{21}}{\hbar}\hat{\mathbf{A}}^{(-)}_{\mathrm{free}}(\mathbf{r}_A,t)\cdot\mathbf{d}_{21}\left(\hat{A}_{22}-\hat{A}_{11}\right), \tag{16}$$

where the rate of spontaneous decay of the excited atomic state $|2\rangle$ is given by

$$\Gamma = \frac{2\omega_A^2 \mu_k \mu_l}{\hbar\epsilon_0 c^2} \,\mathrm{Im}\, G_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A), \tag{17}$$

and the Lamb shift contribution reads

$$\delta\omega = \frac{2\omega_A^2 \mu_k \mu_l}{\hbar\epsilon_0 c^2} \left[\mathrm{Re}\, G_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A) - \frac{1}{\pi} \int_0^\infty d\omega \, \frac{\mathrm{Im}\, G_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega)}{\omega + \omega_A} \right] \tag{18}$$

$[\mu_k \equiv (d_{21})_k$, $\omega_A \equiv \omega_{21}]$. Note that the coincidence limit of the real part of the vacuum Green function is infinite and regularization is required. Here and in the following the vacuum Lamb shift is thought of as being included in the atomic transition frequency, so that $\delta\omega$ only results from the medium. With respect to Green functions of the type given in Eq. (21), with $G^V_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A)$ being the vacuum Green function, $G_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A)$ in Eq. (18) can therefore be replaced with $R_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A)$, and Eq. (17) reduces to

$$\Gamma = \Gamma_0 + \frac{2\omega_A^2 \mu_k \mu_l}{\hbar\epsilon_0 c^2} \,\mathrm{Im}\, R_{kl}(\mathbf{r}_A, \mathbf{r}_A, \omega_A), \tag{19}$$

where $\Gamma_0 = \omega_A^3 \mu^2 / (3\pi\hbar\epsilon_0 c^3)$ is the spontaneous emission rate of the atom in free space.

Equations (17) and (18) hold for all linear dielectrics and arbitrary geometries. Note, that Eq. (17) can be rewritten, on using the relation (10), as

$$\mathrm{Im}\, G_{kk'}(\mathbf{r}, \mathbf{r}', \omega)\delta(\omega - \omega') = \frac{\pi\epsilon_0 c^2}{\hbar\omega^2} \langle 0 | \left[\underline{\hat{E}}_k(\mathbf{r}, \omega), \underline{\hat{E}}^\dagger_{k'}(\mathbf{r}', \omega') \right] |0\rangle, \tag{20}$$

which is nothing but a consequence of the consistent introduction of the noise polarization according to the dissipation-fluctuation theorem. It should be pointed out that for an atom in vacuum the imaginary part of the Green function for arbitrary geometries of the surrounding dielectric is purely transverse and, most importantly, finite.

3 Planar interface

Let us first consider the spontaneous decay of an excited atom near an absorbing planar dielectric surface. This configuration with real refractive index has been studied extensively in the literature in connection with Casimir and van der Waals forces (see [5] and references therein) as well as applications in scanning near-field optical microscopy (SNOM) (see [6] and references therein). To be more specific, we consider two infinite half-spaces (volumes $\mathcal{V}_1$ and $\mathcal{V}_2$) with a common interface such that $\epsilon(\mathbf{r}, \omega) = 1$ if $\mathbf{r} \in \mathcal{V}_1$, and $\epsilon(\mathbf{r}, \omega) = \epsilon(\omega)$ if $\mathbf{r} \in \mathcal{V}_2$ and assume that the atom is placed in $\mathcal{V}_1$ at a distance z from the interface. The Green function in $\mathcal{V}_1$ for this configuration can be given by [2]

$$G_{kk'}(\mathbf{r}, \mathbf{r}', \omega) = G^V_{kk'}(\mathbf{r}, \mathbf{r}', \omega) + R_{kk'}(\mathbf{r}, \mathbf{r}', \omega) \quad (\mathbf{r}, \mathbf{r}' \in \mathcal{V}_1), \tag{21}$$

where $G^V_{kk'}(\mathbf{r}, \mathbf{r}', \omega)$ is the vacuum Green function, and $R_{kk'}(\mathbf{r}, \mathbf{r}', \omega)$ is the reflection Green function. As already mentioned, for calculating the decay rate and the line shift we only need the reflection Green function with both spatial arguments at the position of the atom, $R_{kk'} \equiv R_{kk'}(\mathbf{r}_A, \mathbf{r}_A, \omega)$. Using the formulas given in [2], we obtain

$$R_{xx} = R_{yy} = -\frac{i}{8\pi q^2} \int_0^\infty dk\, k\beta\, \mathrm{e}^{2i\beta z} r^p(k) + \frac{i}{8\pi} \int_0^\infty dk\, \frac{k e^{2i\beta z}}{\beta}\, r^s(k), \tag{22}$$

$$R_{zz} = \frac{i}{4\pi q^2} \int_0^\infty dk\, k^3\, \frac{e^{2i\beta z}}{\beta} r^p(k) \tag{23}$$

($q = \omega/c$, $\beta = \sqrt{q^2 - k^2}$). The functions $r^p(k)$ and $r^s(k)$ are the usual Fresnel reflection coefficients for p- and s-polarized waves. The integrals can be evaluated asymptotically in the case when $qz \ll 1$, i.e. when the distance of the atom from the surface is smaller than the transition wavelength, which is in agreement with the Markov approximation. Expanding in powers of z leads to [$\epsilon \equiv \epsilon(\omega)$, $n \equiv \sqrt{\epsilon(\omega)}$]

$$R_{zz} = \frac{1}{16\pi q^2 z^3} \frac{n^2-1}{n^2+1} + \frac{1}{8\pi z} \frac{(n-1)^2}{n(n+1)} + \frac{iq}{12\pi} \frac{(n-1)(2n-1)}{n(n+1)} + \mathcal{O}(z), \tag{24}$$

$$R_{xx} = R_{yy} = \frac{1}{2} R_{zz} - \frac{1}{16\pi z} \frac{n^2-1}{n^2+1} - \frac{iq}{3\pi} \frac{n-1}{n+1} + \mathcal{O}(z). \tag{25}$$

Inserting Eqs. (24) and (25) into Eq. (19) yields

$$\Gamma = \Gamma_0 \left(1 + \frac{\mu_z^2}{\mu^2}\right) \frac{3}{8(qz)^3} \frac{\epsilon_I(\omega_A)}{|\epsilon(\omega_A)+1|^2} + \mathcal{O}(z^{-1}). \tag{26}$$

The term proportional to z^{-3} exactly agrees with the formula for the leading term derived in [7] in the framework of a microscopic approach, after lengthy and involved diagonalization of the model Hamiltonian used and computation of perturbation series up to second order. It is worth noting that the phenomenologically introduced permittivity and the Green function that is based on it already contain the relevant information, so that the effort is reduced drastically. Needless to say that the microscopic treatment given in [7] is far from a genuine *ab initio* calculation.

SNOM detects surface corrugation or impurities via the changes in the line shift and the line width of an atomic transition far from medium resonances [6]. The high sensitivity of the method results from the cubic dependence of the line shift on the inverse distance of the probe atom from the surface. Disregarding absorption the line width is weakly distance-dependent [cf. Eqs. (19) and (24)], and the strong dependence on distance of the line shift is essentially determined by the first term in Eq. (18),

$$\delta\omega = \Gamma_0 \left(1 + \frac{\mu_z^2}{\mu^2}\right) \frac{3}{4(qz)^3} \frac{\epsilon_R(\omega_A)-1}{\epsilon_R(\omega_A)+1} + \mathcal{O}(z^{-1}) \qquad (\epsilon_I \equiv 0). \tag{27}$$

The situation changes drastically in an absorption band or in the vicinity of it. From Eq. (26) it is seen, that in an absorbing regime the line width also becomes proportional to the inverse cube of the distance. Moreover, the line shift can substantially increase.

To demonstrate the possibility of improving the resolution of SNOM near a medium resonance, we have calculated numerically the spontaneous decay rate as well as the line shift using the single-resonance model permittivity

$$\epsilon(\omega) = 1 + \frac{(0.46\omega_T)^2}{\omega_T^2 - \omega^2 - i\gamma\omega}. \tag{28}$$

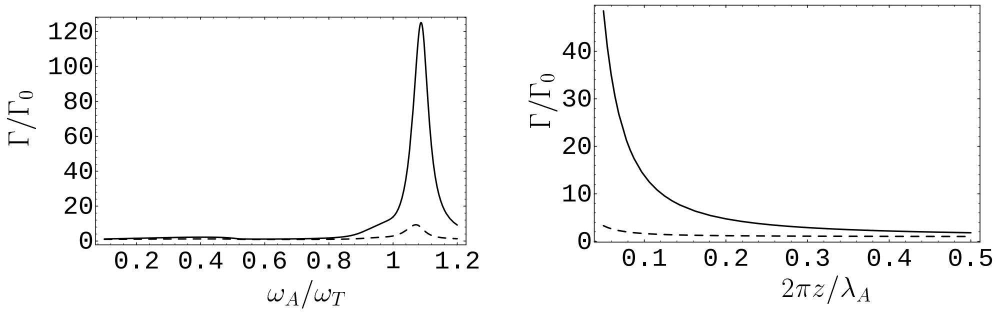

Fig. 1. Decay rate Γ/Γ_0 ($\mu = \mu_z$) for $\gamma/\omega_A = 0.05$ [$\kappa(\omega_A) = 1.29$] as a function of frequency (left figure) for $2\pi z/\lambda_A = 0.1$ (full line) and $2\pi z/\lambda_A = 0.3$ (dashed line), and as a function of distance (right figure) for $\omega_A/\omega_T = 1$ (full line) and $\omega_A/\omega_T = 0.5$ (dashed line).

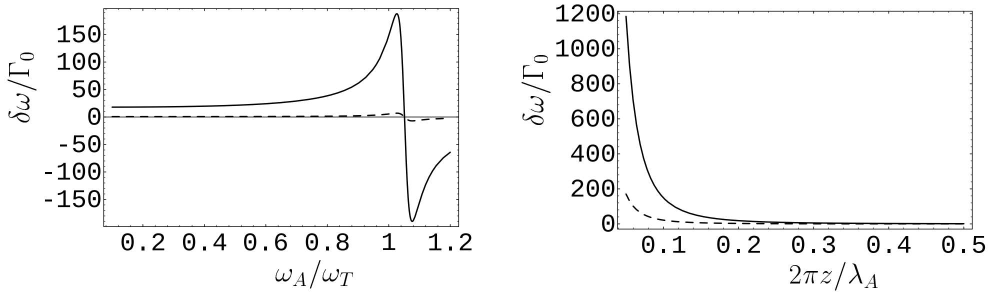

Fig. 2. Line shift $\delta\omega/\Gamma_0$ ($\mu = \mu_z$) for $\gamma/\omega_A = 0.05$ [$\kappa(\omega_A) = 1.29$] as a function of frequency (left figure) for $2\pi z/\lambda_A = 0.1$ (full line) and $2\pi z/\lambda_A = 0.3$ (dashed line), and as a function of distance (right figure) for $\omega_A/\omega_T = 1$ (full line) and $\omega_A/\omega_T = 0.5$ (dashed line).

From Figs. 1 and 2 one observes that near the medium resonance both the decay rate and the line shift exceed substantially the values realized in standard SNOM. Moreover, the resolution of scanning is improved, as can be seen from the vertical line-width resolution

$$\frac{\delta\Gamma}{\delta z} \propto \frac{1}{z^4} \frac{\epsilon_I(\omega_A)}{|\epsilon(\omega_A) + 1|^2} \tag{29}$$

(a similar formula holds for the vertical line-shift resolution). Hence we are left with the proposal of operating in an absorbing regime near a medium resonance in order to improve the sensitivity of measurement.

4 An excited atom in a spherical cavity

Another interesting geometry is the spherical one. In particular, an atom in an empty sphere $\mathcal{V}_R$ of radius R inside an otherwise continuous dielectric medium can serve, in a sense, as a model for spontaneous decay in media (real-cavity model). For $\mathbf{r}, \mathbf{r}' \in \mathcal{V}_R$ the Green function can again be written in the form of Eq. (21), where the reflection tensor reads in dyadic notation as [8]

$$\boldsymbol{R}(\mathbf{r},\mathbf{r}',\omega) = \frac{i\omega}{4\pi c}\sum_{e,o}\sum_{n=1}^{\infty}\sum_{m=0}^{n}\left\{\frac{2n+1}{n(n+1)}\frac{(n-m)!}{(n+m)!}(2-\delta_{0m})\right.$$
$$\left.\times\left[C_n^M(\omega)\boldsymbol{M}_{{}^e_o nm}\!\left(\mathbf{r},\frac{\omega}{c}\right)\boldsymbol{M}_{{}^e_o nm}\!\left(\mathbf{r}',\frac{\omega}{c}\right)+C_n^N(\omega)\boldsymbol{N}_{{}^e_o nm}\!\left(\mathbf{r},\frac{\omega}{c}\right)\boldsymbol{N}_{{}^e_o nm}\!\left(\mathbf{r}',\frac{\omega}{c}\right)\right]\right\}. \quad (30)$$

Here, $\boldsymbol{M}_{{}^e_o nm}(\mathbf{r},k)$ and $\boldsymbol{N}_{{}^e_o nm}(\mathbf{r},k)$ are the ($e$ven and $o$dd) vector Debye potentials, and the quantities $C_n^{M,N}(\omega)$ are the generalized reflection coefficients. Combining Eqs. (19) and (30) (for $\mathbf{r}=\mathbf{r}'=\mathbf{r}_A$), after some algebra we derive

$$\Gamma = \left[1+\mathrm{Re}\,C_1^N(\omega_A)\right]\Gamma_0, \quad (31)$$

where

$$C_1^N(\omega) = \frac{\left[i+z(n+1)-iz^2n-z^3n^2/(n+1)\right]e^{iz}}{\sin z - z(\cos z + in\sin z) + iz^2 n\cos z - z^3(\cos z - in\sin z)n^2/(n^2-1)} \quad (32)$$

($z=R\omega/c$; for details, see [4]). Examples are shown in Figs. 3 and 4. Note that the decay rate is solely related to the transverse electromagnetic field, $\Gamma=\Gamma^{\perp}$.

Restricting our attention to small cavities, $\omega_A R/c\ll 1$, expansion yields

$$\Gamma = \left\{\frac{9\epsilon_I(\omega_A)}{|2\epsilon(\omega_A)+1|^2}\left(\frac{c}{\omega_A R}\right)^3 + \frac{9\epsilon_I(\omega_A)\left[28|\epsilon(\omega_A)|^2+12\epsilon_R(\omega_A)+1\right]}{5|2\epsilon(\omega_A)+1|^4}\left(\frac{c}{\omega_A R}\right)\right.$$
$$+\frac{9\eta(\omega_A)}{|2\epsilon(\omega_A)+1|^4}\left[4|\epsilon(\omega_A)|^4+4\epsilon_R(\omega_A)|\epsilon(\omega_A)|^2+\epsilon_R^2(\omega_A)-\epsilon_I^2(\omega_A)\right]$$
$$\left.-\frac{9\kappa(\omega_A)\epsilon_I(\omega_A)}{|2\epsilon(\omega_A)+1|^4}\left[4|\epsilon(\omega_A)|^2+2\epsilon_R(\omega_A)\right]\right\}\Gamma_0+\mathcal{O}(R) \quad (33)$$

($\epsilon_R\equiv\mathrm{Re}\,\epsilon$, $\epsilon_I\equiv\mathrm{Im}\,\epsilon$). When absorption is fully disregarded, i.e., $\mathrm{Im}\,\epsilon\equiv 0$, Eq. (33) reduces exactly to the well-known Glauber-Lewenstein formula [9]

$$\Gamma = \left(\frac{3n^2}{2n^2+1}\right)^2 n\Gamma_0\,. \quad (34)$$

Needless to say that when setting $\epsilon\equiv 1$, then the free-space spontaneous emission rate is recovered. In absorbing media the decay rate sensitively depends on the cavity radius. Note that the terms $\sim R^{-3}$ and $\sim R^{-1}$ result from absorption. In particular, the term $\sim R^{-3}$ can be regarded as describing nonradiative energy transfer from the guest atom to the constituents of the dielectric via dipole-dipole interaction. It should be pointed out that the above derived rate formula for absorbing media strongly contradicts the rate formula suggested in [10].

5 Virtual-cavity model of spontaneous decay

In the (Clausius-Mosotti) virtual-cavity model it is assumed that the field outside the small sphere is not modified by the sphere, and the local field inside is given by the (smeared) macroscopic field corrected by adding a contribution from the polarization of the sphere. In quantum theory, one has to be careful with the definition of the local field, because of the nonvanishing noise polarization even in the zero-temperature limit. In particular, simple multiplication of the macroscopic electric field with a correction factor as suggested in [10,11] leads to a wrong expression for the rate of spontaneous decay and misinterpretations concerning the action of transverse and longitudinal fields. Introducing the local field as $\underline{\hat{\mathbf{E}}}'(\mathbf{r},\omega) = \underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) + \underline{\hat{\mathbf{P}}}(\mathbf{r},\omega)/(3\epsilon_0)$, from Eqs. (2) and (4) together with Eqs. (5) – (7) it follows that [4,12]

$$\underline{\hat{\mathbf{E}}}'(\mathbf{r},\omega) = \frac{1}{3}\big[\epsilon(\mathbf{r},\omega)+2\big]\underline{\hat{\mathbf{E}}}(\mathbf{r},\omega) + \frac{1}{3\epsilon_0}\underline{\hat{\mathbf{P}}}^N(\mathbf{r},\omega), \tag{35}$$

with

$$\underline{\hat{\mathbf{P}}}^N(\mathbf{r},\omega) = -\frac{1}{i\omega}\underline{\hat{\mathbf{j}}}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar\epsilon_0}{\pi}\,\epsilon_I(\mathbf{r},\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega). \tag{36}$$

The spontaneous decay rate can now be calculated using Eqs. (17) and (20) together with the local field defined in Eq. (35) and the Green function for bulk material, $G^M_{kk'}(\mathbf{r},\mathbf{r}',\omega_A)$. However, for $\mathbf{r},\mathbf{r}'\to\mathbf{r}_A$ the imaginary part of $G^M_{kk'}(\mathbf{r},\mathbf{r}',\omega_A)$ becomes singular, which reflects the fact that the dielectric is thought of as being smeared out even over the region where the atom is located. In order to overcome this difficulty, regularization is required. A small distance between the spatial arguments $\mathbf{r}$ and $\mathbf{r}'$ can be kept, and the decay rate can be obtained by averaging over an appropriately chosen sphere of radius R (virtual cavity),

$$\begin{aligned}\Gamma \;=\;& \frac{2\omega_A^2\mu_k\mu_{k'}}{\hbar\epsilon_0c^2}\left|\frac{\epsilon(\omega_A)+2}{3}\right|^2 \operatorname{Im}\overline{G^M_{kk'}(\mathbf{r},\mathbf{r}',\omega_A)} + \frac{2\mu_k\mu_{k'}}{9\hbar\epsilon_0}\epsilon_I(\omega_A)\,\overline{\delta_{kk'}\delta(\mathbf{r}-\mathbf{r}')}\\ &+\frac{4\omega_A^2\mu_k\mu_{k'}}{3\hbar\epsilon_0c^2}\,\epsilon_I(\omega_A)\operatorname{Re}\left[\frac{\epsilon(\omega_A)+2}{3}\,\overline{G^M_{kk'}(\mathbf{r},\mathbf{r}',\omega_A)}\right]\end{aligned} \tag{37}$$

(for details, see [4]). Obviously, different averaging procedures leads to (slightly) different numerical prefactors in the R-dependent terms [4,12].

Let us consider a small cavity ($|R\omega_A n/c| \ll 1$). Averaging with respect to both spatial arguments separately over the sphere with equal weight, we obtain [12]

$$\Gamma = \Gamma^\perp + \Gamma^\parallel, \tag{38}$$

$$\begin{aligned}\Gamma^\perp = \Bigg\{&\frac{25\epsilon_I(\omega_A)}{54}\left(\frac{c}{\omega_AR}\right)^3 + \epsilon_I(\omega_A)\left[\epsilon_R(\omega_A)+2\right]\left[\frac{8}{15}\left(\frac{c}{\omega_AR}\right) - \frac{2\kappa(\omega_A)}{9}\right]\\ &+\eta(\omega_A)\left[\left|\frac{\epsilon(\omega_A)+2}{3}\right|^2 - \frac{2\epsilon_I^2(\omega_A)}{9}\right]\Bigg\}\Gamma_0 + \mathcal{O}(R),\end{aligned} \tag{39}$$

$$\Gamma^\parallel = \Gamma_0\frac{4\epsilon_I(\omega_A)}{27|\epsilon(\omega_A)|^2}\left(\frac{c}{\omega_AR}\right)^3. \tag{40}$$

Here, $\Gamma^{\perp}$ and $\Gamma^{\|}$, respectively, are related to the transverse and longitudinal parts of the electromagnetic fields. It should be pointed out that the appearance of $\Gamma^{\|}$ is due to the fact that (in contrast of the real-cavity model) the atom is not strictly located in vacuum and the imaginary part of the longitudinal Green function (at equal space points) does not vanish for absorbing media.

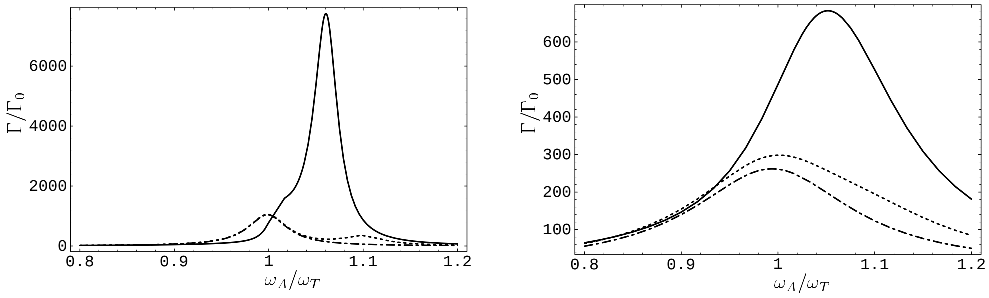

Fig. 3. Spontaneous decay rate Γ/Γ_0 for $R=0.02\lambda_A$ and $\gamma=0.05\omega_T$ (left figure) and $\gamma=0.2\omega_T$ (right figure). The solid lines correspond to the real-cavity model, Eq. (33), and the dotted lines correspond to the virtual-cavity model, Eqs. (39) and (40), with the broken line indicating the transverse part (39).

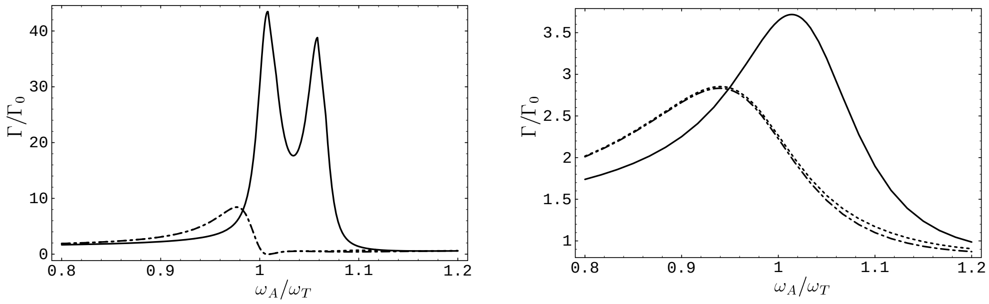

Fig. 4. Spontaneous decay rate Γ/Γ_0 for $R=0.2\lambda_A$ and $\gamma=0.05\omega_T$ (left figure) and $\gamma=0.2\omega_T$ (right figure). The solid lines correspond to the real-cavity model, Eq. (33), and the dotted lines correspond to the virtual-cavity model, Eqs. (39) and (40), with the broken line indicating the transverse part (39).

Examples of Γ and $\Gamma^{\perp}$ are shown in Figs. 3 and 4. The results differ from those in [10,11] in the contribution of the noise polarization $\hat{\mathbf{P}}^{N}$, which is disregarded in [10,11]. It is worth noting that the effect of the noise polarization has recently been fully confirmed on the basis of a microscopic approach [13]. The effect vanishes for nonabsorbing media ($\epsilon_I \equiv 0$), and Eq. (39) yields the well-known rate formula

$$\Gamma = \Gamma^{\perp} = \left(\frac{n^2+2}{3}\right)^2 n\Gamma_0. \tag{41}$$

6 Summary

We have shown how quantization of the phenomenological electrodynamics in linear media can be most easily employed to study radiating atoms in the presence of dispersive and absorbing dielectric bodies and to calculate the line widths and line shifts of the atomic transitions. It is worth noting that the approach reproduces typical results that have been derived using quite different concepts including involved microscopic ones. The approach also shows that the rate formulas given in [10,11] for the spontaneous decay of excited atoms in absorbing media are not correct, because they are based on wrong respective incomplete medium-assisted vacuum noise. In particular, we have considered a planar interface and a spherical cavity, which may serve as a model of spontaneous decay in media, and we have compared it with the corresponding (Clausius-Mosotti) virtual-cavity model. The results reveal that when an excited atom approaches an absorbing dielectric body, then the rate of spontaneous decay becomes proportional to the inverse cubic distance of the atom from dielectric body [see, e.g., Eqs. (26) and (33)]. In microscopic models, terms of that type have been shown to correspond to nonradiative energy transfer from the atom to the medium via dipole-dipole interaction, i.e., a second-order process with no photon in the final state. With regard to applications, the results show that absorption offers the possibility of improving the resolution of SNOM.

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