Quantized Media with Absorptive Scatterers and Modified Atomic Emission Rates

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Abstract

Modifications in the spontaneous emission rate of an excited atom that are caused by extinction effects in a nearby dielectric medium are analyzed in a quantummechanical model, in which the medium consists of spherical scatterers with absorptive properties. Use of the dyadic Green function of the electromagnetic field near a dielectric sphere leads to an expression for the change in the emission rate as a series of multipole contributions for which analytical formulas are obtained. The results for the modified emission rate as a function of the distance between the excited atom and the dielectric medium show the influence of both absorption and scattering processes.

1. Introduction

The emission rate of an excited atom is modified if the electromagnetic properties of its surroundings differ from that of vacuum [1]. For an atom in front of a dielectric medium filling a half-space, the rate varies with the distance between the atom and the medium [2 - 8]. Usually, the medium is taken to be homogeneous on the scale of the atomic wavelength, so that its electromagnetic properties are fully described by a susceptibility, which does not vary appreciably on the scale of the wavelength. In general, it will be complex to account for absorption and dispersion. For such a configuration modifications of the atomic radiative properties have been confirmed experimentally a few years ago [9].

If the structure of the medium cannot be neglected, scattering effects play a role as well, so that extinction in such a medium is driven by both absorption and scattering. Extinction by scattering in material media is quite common, owing to the presence of impurities and defects. The interplay between the two types of extinction in atomic decay rates can be investigated in a model in which both of these features occur simultaneously. In a recent paper [10] we have studied the change in the decay rate of an atom in the presence of a medium consisting of non-overlapping spheres that are made of absorptive dielectric material. The spheres are distributed randomly in a half-space, with a uniform average density. In order to describe the absorptive dielectric material of the spheres in a quantummechanically consistent way a damped-polariton model has been employed [11]. By introducing an effective susceptibility for the composite medium and after a detailed analysis of surface contributions, we could derive an asymptotic expression for the change in the emission rate at relatively large distances between the atom and the medium.

In the present paper we take a somewhat different approach so as to derive an analytic expression for the emission rate that is valid for all distances between the atom and the medium. We shall start from exact expressions for the electromagnetic Green function in the presence of a dielectric sphere of arbitrary radius. As before, the absorptive dielectric material of the spheres will be described by a damped-polariton model. For simplicity we shall assume that the density of the spherical scatterers in the medium is low, so that multiple-scattering effects can be neglected.

2. Spontaneous emission in the presence of absorbing dielectrics

In the damped-polariton model an absorptive linear dielectric medium is described by a polarization density that is coupled to a bath of harmonic oscillators with a continuous range of frequencies [11]. The Hamiltonian of the damped-polariton model can be diagonalized exactly, as has been shown both for the case of a uniform dielectric [11] and for a dielectric with arbitrary inhomogeneities [12]. Diagonalization of the Hamiltonian in the general non-homogeneous case yields

\[ H_d = \int dr \int_0^\infty d\omega \hbar \omega C^\dagger(r, \omega) \cdot C(r, \omega), \]  

(2.1)

with annihilation operators \( C(r, \omega) \) and associated creation operators. The electric field can be expressed in terms of these operators as [12]:

\[ E(r) = \int dr' \int_0^\infty d\omega f_E(r, r', \omega) \cdot C(r', \omega) + \text{h.c.}, \]  

(2.2)

with a tensorial coefficient:

\[ f_E(r, r', \omega) = -\frac{\omega^2}{c^2} \frac{\hbar \text{Im} \epsilon(r', \omega + i\delta)}{\pi \epsilon_0} \left( \frac{\epsilon(r, \omega + i\delta)}{\pi \epsilon_0} \right)^{1/2} G(r, r', \omega + i\delta). \]  

(2.3)

Here \( \epsilon \) is the complex local (relative) dielectric constant, which follows from the parameters of the model. Furthermore, \( G \) is the tensorial Green function, which satisfies the differential equation

\[ -\nabla \times [\nabla \times G(r, r', \omega + i\delta)] + \frac{\omega^2}{c^2} \epsilon(r, \omega + i\delta) G(r, r', \omega + i\delta) = 1 \delta(r - r'). \]  

(2.4)
with $\mathbf{I}$ the unit tensor.

The atomic decay rate in the presence of an absorbing dielectric follows from the inhomogeneous damped-polariton model in its diagonalized form by employing perturbation theory in leading order \cite{10}. It can be expressed as an integral over a product of the coefficients (2.3) and suitable atomic matrix elements:

$$\Gamma = \frac{2\pi}{h^2\omega_o^2} \int dr \int dr' \int d\mathbf{r} \int d\mathbf{r}' \int d\mathbf{r}'' \langle \mathbf{J}_a(r') | g | \mathbf{I}_e(r', r, \omega_o) \rangle \cdot \mathbf{F}_e(r'', r, \omega_o) \cdot \langle \mathbf{J}_a(r'') | e \rangle \cdot \langle \mathbf{J}_a(r'') | g | \mathbf{I}_e(r, r', \omega_o) \rangle \cdot \mathbf{F}_e(r, r', \omega_o) \cdot \langle \mathbf{J}_a(r) | e \rangle \cdot \langle \mathbf{J}_a(r) | g | \mathbf{I}_e(r, r, \omega_o) \rangle \cdot \mathbf{F}_e(r, r, \omega_o) \cdot \langle \mathbf{J}_a(r) | e \rangle,$$

(2.5)

with $e$ and $g$ denoting the excited and the ground state of the atom, $\omega_o$ the atomic transition frequency, and the tilde denoting the tensor transpose. Furthermore, $\mathbf{J}_a(r)$ is the atomic local current density $-\frac{i}{\hbar} e \sum_j \langle \mathbf{p}_j/m, \delta(\mathbf{r}-\mathbf{r}_j) \rangle$, with $\mathbf{r}_j, \mathbf{p}_j$ the positions and momenta of the electrons and curly brackets denoting the anti-commutator. In the electric-dipole approximation the atomic decay rate can be expressed in terms of the Green function as:

$$\Gamma = \frac{2\hbar^2}{\omega_o^2} \langle e | \mathbf{d} | g \rangle \cdot \text{Im} \sum_{\ell,\ell'} \sum_{m=-\ell}^{\ell} \frac{(-1)^{m}}{\ell+1} \cdot \langle \mathbf{J}_a(r') | \mathbf{I}_e(r', r, \omega_o + i0) \rangle \cdot \langle g | \mathbf{I}_e(r, r', \omega_o) \rangle\cdot \langle \mathbf{J}_a(r) | e \rangle \cdot \langle e | \mathbf{d} | g \rangle \cdot \text{Im} \sum_{\ell,\ell'} \sum_{m=-\ell}^{\ell} \frac{(-1)^{m}}{\ell+1} \cdot \langle \mathbf{J}_a(r') | \mathbf{I}_e(r', r, \omega_o + i0) \rangle \cdot \langle g | \mathbf{I}_e(r, r', \omega_o) \rangle\cdot \langle \mathbf{J}_a(r) | e \rangle,$$

(2.6)

with $r_0$, the atomic position and $\mathbf{d} = -e \sum_j \langle \mathbf{r}_j - \mathbf{r}_0 \rangle$ the atomic electric dipole moment. The above expression for the decay rate of an excited atom in the presence of an inhomogeneous absorptive dielectric can be obtained as well by invoking the fluctuation-dissipation theorem \cite{13,14}.

3. Green functions

The Green function in vacuum fulfills the differential equation (2.2) with $\omega = 1$. It follows from the scalar Green function $G_s(\mathbf{r}, \mathbf{r}', \omega) = \exp(\mathrm{i} \omega r - r'/c)/(4\pi r - r')$ as $G_0(\mathbf{r}, \mathbf{r}', \omega) = -1 + (c^2/\omega^2) \nabla \nabla G_s(\mathbf{r}, \mathbf{r}', \omega)$. Its explicit form in spherical coordinates is obtained from the expansion of $G_s$ in spherical harmonics and spherical Bessel functions. The ensuing form for the vacuum Green function is \cite{15,16}:

$$G_0(\mathbf{r}, \mathbf{r}', \omega + i0) = \frac{-ik}{8\pi kr} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{(-1)^m}{\ell+1} \frac{1}{\ell+1} \left[ \left( M_{\ell,m}(r) Y_{\ell,m}(r') + N_{\ell,m}(r) Y_{\ell,m}(r') \right) \right]$$

$$+ \theta(\mathbf{r} - \mathbf{r}') \left[ \left( M_{\ell,m}(r) Y_{\ell,m}(r') + N_{\ell,m}(r) Y_{\ell,m}(r') \right) \right]$$

$$+ k^2 \mathbf{e}_e \cdot \mathbf{e}_e \cdot \delta(\mathbf{r} - \mathbf{r}'),$$

(3.1)

with $k = \omega/c$ the wavenumber, $\mathbf{e}_e$ a unit vector in the direction of $\mathbf{r}$ and $\theta(\mathbf{r})$ a step function that equals 1 for positive and 0 for negative argument. The vector harmonics are defined as:

$$M_{\ell,m}(r) = \nabla \wedge [\mathbf{r} Y_{\ell,m}(r)],$$

(3.2)

$$N_{\ell,m}(r) = k^{-1} \nabla \wedge [\nabla \wedge [\mathbf{r} Y_{\ell,m}(r)]],$$

(3.3)

where $Y_{\ell,m}(r)$ stands for $j_{\ell}(kr) Y_{\ell,m}(\theta, \phi)$, with $j_{\ell}$ spherical Bessel functions and $Y_{\ell,m}$ spherical harmonics. The superscripts $(h)$ in (3.1) denote the analogous vector harmonics with spherical Hankel functions $h_{\ell}^{(1)}(r)$ instead of $j_{\ell}(r)$.

The expression (3.1) may be checked by substitution in the differential equation (2.4). Differentiation of the step functions yields singular terms, which together with the last term lead to the right-hand side of (2.4).

The Green function in the presence of a dielectric sphere is the sum of the vacuum Green function and a correction term. For a sphere centered at the origin the latter has the form \cite{15,17}:

$$G_s(\mathbf{r}, \mathbf{r}', \omega + i0) = \frac{k}{8\pi kr} \sum_{\ell=1}^{\infty} \sum_{m=-\ell}^{\ell} \frac{(-1)^m}{\ell+1} \frac{1}{\ell+1} \left[ \left( B_{\ell}^o N_{\ell,m}^{(b)}(r') Y_{\ell,m}(r) + B_{\ell}^o M_{\ell,m}^{(b)}(r) N_{\ell,m}^{(b)}(r) \right) \right]$$

(3.4)

for $\mathbf{r}$ and $\mathbf{r}'$ both outside the sphere. The electric and magnetic multipole amplitudes read \cite{18-20}:

$$B_{\ell}^o = \frac{i^{\ell+1}}{\ell+1} \frac{N_{\ell}^{0}}{\ell(\ell+1)} D_{\ell}^o,$$

(3.5)

with $p = e, m$. The numerators and denominators are given as:

$$N_{\ell}^e = e \times f_{\ell}(q) j_{\ell}(q') - j_{\ell}(q) f_{\ell}(q'),$$

$$N_{\ell}^m = f_{\ell}(q) j_{\ell}(q') - j_{\ell}(q) f_{\ell}(q'),$$

$$D_{\ell}^e = e \times f_{\ell}(q) j_{\ell}(q') - h_{\ell}^{(1)}(q) f_{\ell}(q'),$$

$$D_{\ell}^m = f_{\ell}(q) j_{\ell}(q') - h_{\ell}^{(1)}(q) f_{\ell}(q'),$$

(3.6)

with $f_{\ell}(q) = (\ell+1) j_{\ell}(q) - q j_{\ell+1}(q)$ and $f_{\ell}^{(b)}(q) = (\ell+1) h_{\ell}^{(1)}(q) - q h_{\ell+1}^{(1)}(q)$. The spherical Bessel and Hankel functions depend on $q = kr$ and $q' = kq$, with $a$ the radius of the sphere.

To determine the change in the atomic decay rate due to the presence of a dielectric sphere one needs the components of the Green function $G_s$ for coinciding arguments. The non-vanishing components follow from (3.4) as:

$$\mathbf{e}_e \cdot G_s(\mathbf{r}, \mathbf{r}, \omega + i0) \cdot \mathbf{e}_e =$$

$$= \frac{1}{4\pi kr} \sum_{\ell=1}^{\infty} \sum_{m=1}^{\infty} \frac{(-1)^m}{\ell+1} \frac{1}{\ell+1} \left[ \left( B_{\ell}^o \left[ h_{\ell}^{(1)}(kr) \right]^2 \right) \right]$$

(3.7)

and

$$\mathbf{e}_e \cdot G_s(\mathbf{r}, \mathbf{r}, \omega + i0) \cdot \mathbf{e}_e = \mathbf{e}_e \cdot G_s(\mathbf{r}, \mathbf{r}, \omega + i0) \cdot \mathbf{e}_e =$$

$$= \frac{1}{8\pi kr^2} \sum_{\ell=1}^{\infty} \sum_{m=1}^{\infty} \frac{(-1)^m}{\ell+1} \frac{1}{\ell+1} \left[ \left( B_{\ell}^o \left[ d/dr \left[ h_{\ell}^{(1)}(kr) \right] \right]^2 \right) + B_{\ell}^o \left[ kr h_{\ell}^{(1)}(kr) \right]^2 \right],$$

(3.8)

in agreement with \cite{21}. Here $\mathbf{e}_e, \mathbf{e}_e$ and $\mathbf{e}_e$ are unit vectors in a spherical coordinate system.

4. Decay near a half-space of absorptive scatterers

We consider a halfspace $z < 0$ filled with a dilute set of spherical scatterers. The non-overlapping spheres are randomly distributed with a uniform average density. An excited atom is located at $\mathbf{r}_0 = (0,0,z_0)$, with $z_0 > a$, so that the minimal distance between the atom and the scatterers is positive. The decay rate is given by the sum of the vacuum decay rate and a correction term. The modified rate depends on the orientation of
the dipole-moment transition matrix element. If the dipole-moment is oriented perpendicular to the $z$-axis, the vacuum rate is $G_{0,0} = \omega_0^2 \langle |\langle e | \mu_\perp | g \rangle |^2 / (3\pi \epsilon_0 hc^3)$. A similar formula is valid for a dipole moment oriented parallel to the $z$-axis, with $\mu_\perp$ replaced by $\mu_z$.

If multiple-scattering effects are neglected, the correction term in the decay rate is given by the sum of the correction terms due to all spheres. For the perpendicular orientation one finds

$$G_{\perp,\perp} = -\frac{2\omega_0^2}{\epsilon_0 hc^2} \sum_j \langle e | \mu_\perp | g \rangle \cdot \Im G_j (r_a - R_a - r_j - R_j, \omega_0 + i0) \cdot (g | \mu_\perp | e),$$  \hspace{1cm} (4.1)

with $G_j$ given by (3.4) and $R_j$ the positions of the centers of the spheres. Choosing the $x$-axis to be parallel to the transition matrix element and averaging over the positions of the spheres we get

$$\langle G_{\perp,\perp} \rangle = \frac{6\pi n e^2}{\omega_0} \Gamma_{0,0} \operatorname{Im} \int_{t_0}^{\infty} dt \times e_x \cdot G_j (r_a - r - r_j - R_j, \omega_0 + i0) \cdot e_x,$$  \hspace{1cm} (4.2)

with $n$ the uniform density of the spheres and $e_x$ a unit vector along the $x$-axis. The volume integral can be written as a triple integral, viz. over $|r - r_j|, z$ and an azimuthal angle. Upon carrying out the latter two of these integrals one finds for the volume integral in (4.2):

$$\pi \int_{z_a}^{\infty} \frac{dz}{z} \left( \frac{3}{2} z_a^2 - z_a r + \frac{r^2}{2} \right) e_y \cdot G_j (r_a - r - r_j - R_j, \omega_0 + i0) \cdot e_y + \left( \frac{3}{2} z_a^2 + \frac{r^2}{2} \right) e_y \cdot G_j (r_a - r - r_j - R_j, \omega_0 + i0) \cdot e_y + \left( -z_a r + \frac{r^2}{2} \right) e_y \cdot G_j (r_a - r - r_j - R_j, \omega_0 + i0) \cdot e_y,$$  \hspace{1cm} (4.3)

Insertion of (3.7) and (3.3) yields

$$\langle G_{\perp,\perp} \rangle = \frac{3\pi n e^2}{4\omega_0} \Gamma_{0,0} \sum_{\ell_1=1}^{\infty} (-i)^{\ell_1} \ell (\ell + 1) \times B_{\ell_1}^m J^{m}_{\ell_1}(\zeta_0) + B_{\ell_1}^m J^{m}_{\ell_1}(\zeta_0),$$  \hspace{1cm} (4.4)

with multipole amplitudes $B_{\ell_1}^m$ given by (3.5)-(3.9) with $k = \omega_0 / c$, and with the integrals

$$J^{m}_{\ell_1}(\zeta_0) = 2\ell (\ell + 1) \int_{\zeta_0}^{\infty} dt \left( \frac{3}{2\pi} - \frac{\zeta}{\ell + 1} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2 + \int_{\zeta_0}^{\infty} dt \left( \frac{3}{2\pi} + \frac{\zeta}{\ell + 1} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2,$$  \hspace{1cm} (4.5)

and

$$J^{m}_{\ell_1}(\zeta_0) = \int_{\zeta_0}^{\infty} dt \left( \frac{3}{2\pi} - \zeta - \frac{\ell_1 + 1}{2} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2,$$  \hspace{1cm} (4.6)

with $\zeta$ equal to $\zeta_a = (\omega_0 + i0)z_a / c$. The derivative of the spherical Hankel function in (4.5) can be rewritten in terms of Hankel functions with a different index [22]. For large $\zeta$ the asymptotic forms of these integrals are

$$J^{m}_{\ell_1}(\zeta) \approx (-1)^{\ell_1+1} \frac{e^{2i\zeta c}}{2\zeta}, \quad J^{m}_{\ell_1}(\zeta) \approx (-1)^{\ell} \frac{e^{2i\zeta c}}{2\zeta},$$  \hspace{1cm} (4.7)

so that (4.4) becomes

$$\langle G_{\perp,\perp} \rangle = \frac{3\pi n e^2}{8\omega_0 c^2} \Gamma_{0,0} \sum_{\ell_1=1}^{\infty} \ell (\ell + 1) (B_{\ell_1}^c - B_{\ell_1}^m) e^{2i\zeta_0},$$  \hspace{1cm} (4.8)

which falls off proportionally to $1/\zeta_0$. Substituting the leading terms of $B_{\ell_1}^c, B_{\ell_1}^m$ and $B_{\ell_1}^m$ for small values of $q$ and $\epsilon - 1$ one recovers a result found before [10].

Similar expressions may be obtained for the correction to the decay rate of an excited atom with a dipole moment parallel to the $z$-axis. Instead of (4.3) one gets a formula with the $z$-component of $G_j$. Upon carrying out the integrals one arrives at the analogue of (4.4), with the integrals

$$J^{c}_{\ell_1}(\zeta) = 2\ell (\ell + 1) \int_{\zeta}^{\infty} dt \left( \frac{3}{2\pi} + \frac{\zeta}{\ell + 1} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2 + \int_{\zeta}^{\infty} dt \left( \frac{3}{2\pi} + \frac{\zeta}{\ell + 1} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2,$$  \hspace{1cm} (4.9)

and

$$J^{m}_{\ell_1}(\zeta) = \int_{\zeta}^{\infty} dt \left( \frac{3}{2\pi} - \zeta + \frac{2\ell + 1}{3} \right) \left| h^{(1)}_{\ell_1}(t) \right|^2.$$  \hspace{1cm} (4.10)

For large $\zeta$ their asymptotic forms are

$$J^{c}_{\ell_1}(\zeta) \approx (-1)^{\ell+1} \frac{e^{2i\zeta c}}{2\zeta}, \quad J^{m}_{\ell_1}(\zeta) \approx (-1)^{\ell} \frac{e^{2i\zeta c}}{2\zeta},$$  \hspace{1cm} (4.11)

so that the decay rate for large $\zeta_0$ becomes

$$\langle G_{c,\perp} \rangle = \frac{3\pi n e^2}{8\omega_0 c^2} \Gamma_{0,0} \sum_{\ell_1=1}^{\infty} \ell (\ell + 1) (B_{\ell_1}^c - B_{\ell_1}^m) e^{2i\zeta_0}.$$  \hspace{1cm} (4.12)

In contrast to (4.8) the right-hand side is proportional to the inverse square of $\zeta_0$. For general values of $\zeta_0$ we have to evaluate the integrals in (4.5)-(4.6) and (4.5)-(4.10), as will be done in the following section.

5. Evaluation of integrals

The integrals $J^{c}_{\ell_1}$ and $J^{m}_{\ell_1}$ are linear combinations of integrals of the general form

$$I_{\ell_1,\ell_2,n}(\zeta) = \int_{1}^{\infty} du u^n h^{(1)}_{\ell_1}(\zeta u) h^{(1)}_{\ell_2}(\zeta u),$$  \hspace{1cm} (5.1)

which is symmetric in $\ell_1, \ell_2$. In fact, upon inspecting (4.5)-(4.6) and (4.9)-(4.10) we find that explicit expressions are needed for the integrals $I_{\ell_1,\ell_2,n}(\zeta)$ with $n = -2, -1, 0, 1, 3$ and for $I_{\ell_1,1,n}(\zeta)$ for $n = -1, 0, 2$. With the use of standard identities [22] for spherical Hankel functions and by means of a partial integration we may derive several relations connecting these integrals for different values of the parameters:

$$I_{\ell_1-1,\ell_2,n}(\zeta) + I_{\ell_1+1,\ell_2,n}(\zeta) = \frac{2\ell_1 + 1}{\zeta} I_{\ell_1,\ell_2,n+1}(\zeta),$$  \hspace{1cm} (5.2)

$$I_{n-\ell_1-\ell_2,n}(\zeta) + (n + \ell_1 - \ell_2 + 1) I_{\ell_1+1,\ell_2,n}(\zeta) + (2\ell_1 + 1) I_{\ell_1,\ell_2+n+1}(\zeta) = \frac{2\ell_1 + 1}{\zeta} I_{\ell_1,\ell_2,n}(\zeta).$$  \hspace{1cm} (5.3)
In order to obtain explicit expressions for \( I_{\ell_1,\ell_2, n} \) with \( n = 0, 1, 3 \) we start from a result \( \text{[5.2]} \) that is valid for \( n = 0 \) and \( \ell_1 \neq \ell_2 \):

\[
(\ell_1 + \ell_2 + 1)I_{\ell_1,\ell_2,0}(\zeta) = \frac{\zeta}{\ell_1 - \ell_2} \left( h^{(1)}_{\ell_1}h^{(1)}_{\ell_2} - h^{(1)}_{\ell_1+1}h^{(1)}_{\ell_2} + h^{(1)}_{\ell_1}h^{(1)}_{\ell_2+1} \right),
\tag{5.4}
\]
as may be checked by differentiation. We omit the argument \( \zeta \) of the spherical Hankel functions from now on. To obtain the corresponding expression for \( \ell_1 = \ell_2 \) we put \( \ell_1 = \ell + 1 \), \( \ell_2 = \ell \) and \( n = 0 \) in \( \text{[5.3]} \) and use \( \text{[5.4]} \) in the second term. In this way we obtain a recursion relation connecting \( I_{\ell_1,\ell_2,0} \) for consecutive values of \( \ell \). Solving this relation by employing the identity \( I_{0,0,0}(\zeta) = -(2i/\zeta)E_1(-2i\zeta) + [h^{(1)}_0]^2 \) (with \( E_1 \) the exponential integral \( \text{[2.2]} \)) as an initial condition, we find

\[
I_{\ell,\ell,0}(\zeta) = -\frac{2i}{(2\ell + 1)^2}E_1(-2i\zeta) + \frac{\ell}{2\ell + 1} \left[ h^{(1)}_\ell \right]^2 \tag{5.5}
\]
for all \( \ell \geq 0 \). The exponential integral of purely imaginary argument can be expressed in terms of sine and cosine integrals as \( E_1(-2i\zeta) = -\text{Ci}(2\zeta) - i\text{Si}(2\zeta) + i\pi/2 \). With the help of the identities \( \text{[5.2]} \), \( \text{[5.4]} \) and the recursion relations for the spherical Hankel functions one derives expressions for \( I_{\ell,\ell,1} \) (with \( \ell \geq 1 \)) and \( I_{\ell,\ell,3} \) (with \( \ell \geq 2 \)), in the form of linear combinations of products of spherical Hankel functions:

\[
I_{\ell,\ell,1}(\zeta) = -\frac{\zeta^2}{2\ell(\ell + 1)} + \left[ h^{(1)}_\ell \right]^2 \tag{5.6}
\]
for \( \ell \geq 1 \), and

\[
I_{\ell,\ell,3}(\zeta) = \frac{\zeta^4}{3(\ell - 1)(\ell + 1)(\ell + 2)} - \frac{\zeta^2}{6(\ell + 1)(\ell + 2)}
+ \frac{\zeta}{2(\ell + 2)} \left[ h^{(1)}_\ell \right]^2 + \frac{\zeta^4}{3(\ell - 1)(\ell + 1)(\ell + 2)}
- \frac{\zeta^2}{6(\ell - 1)(\ell + 2)} \left[ h^{(1)}_{\ell-1} \right]^2 + \frac{2\zeta^3}{3(\ell - 1)(\ell + 1)(\ell + 2)}
+ \frac{\zeta}{3(\ell + 2)} \left[ h^{(1)}_{\ell}h^{(1)}_{\ell-1} \right] \tag{5.7}
\]
for \( \ell \geq 2 \). It turns out that these formulas cannot be used for \( I_{0,0,1} \) and \( I_{1,1,3} \), since the expressions diverge in these cases. However, these special cases can be obtained straightforwardly by connecting them to the exponential integral of the same argument as in \( \text{[5.5]} \).

Expressions for \( I_{\ell_1,\ell_2, n} \) with \( n = -2 \) follow by choosing \( \ell_1 = \ell + 1 \), \( \ell_2 = \ell \) and \( n = -2 \) in \( \text{[5.3]} \). The second term at the left-hand side drops out for these values of the parameters. As a result a simple recurrence relation for \( I_{\ell_1,\ell_2, -2} \) is found, which may be solved for all \( \ell \geq 0 \) by employing the identity \( I_{0,0,-2}(\zeta) = -ie^{2\zeta^2/(2\zeta^2)} \) as a starting point. One gets for \( \ell \geq 0 \):

\[
I_{\ell_1,\ell_2,-2}(\zeta) = -\frac{1}{2} \left[ h^{(1)}_{\ell} \right]^2 - \frac{1}{2} \left[ h^{(1)}_{\ell+1} \right]^2 + \frac{2\zeta + 1}{2\zeta} h^{(1)}_{\ell}h^{(1)}_{\ell+1} \tag{5.8}
\]
Furthermore, by choosing in \( \text{[5.3]} \) the parameters as \( n = -2 \) and \( \ell_1 = \ell_2 \) as either \( \ell \) or \( \ell + 1 \), one gets two identities, which may be combined with \( \text{[5.2]} \) so as to obtain a recursion relation for \( I_{\ell_1,\ell_2, -1} \). Solving that relation with the initial condition \( I_{0,0,-1}(\zeta) = -\zeta^2E_1(-2i\zeta) \), we find for all \( \ell \geq 0 \):

\[
I_{\ell_1,\ell_2,-1}(\zeta) = -\zeta^2E_1(-2i\zeta) + \sum_{k=1}^\ell \frac{2k + 1}{2(k + 1)} \left[ h^{(1)}_{k} \right]^2
+ \frac{1}{2} \left[ h^{(1)}_0 \right]^2 - \frac{1}{2\ell(\ell + 1)} \left[ h^{(1)}_{\ell} \right]^2 \tag{5.9}
\]
It should be noted that the sum drops out for \( \ell = 0 \).

Finally, we need expressions for \( I_{\ell_1,\ell_2,-p} \) for \( p = -1 \) and \( p = 2 \). Once more we use the identity \( \text{[5.3]} \), now for the choice \( \ell_1 = \ell_2 = \ell \) and \( n = -1 \). It yields a recursion relation for \( I_{\ell_1,\ell_2,-1} \) from which we get for \( \ell \geq 1 \):

\[
I_{\ell_1,\ell_2,-1}(\zeta) = -i\zeta^2E_1(-2i\zeta) + \zeta^3 \sum_{k=0}^{\ell-1} \left[ h^{(1)}_k \right]^2 \tag{5.10}
\]
Turning to the case \( p = 2 \), one derives a result for \( I_{\ell_1,\ell_2,2} \) by a repeated use of \( \text{[5.2]} \) in combination with \( \text{[5.4]} \). Once again linear combinations of products of two spherical Hankel functions are found, at least for \( \ell \geq 2 \):

\[
I_{\ell_1,\ell_2,2}(\zeta) = \left[ -\frac{\zeta^3}{3(\ell - 1)(\ell + 1)} - \frac{\zeta^2}{6(\ell + 1)} \right] \left[ h^{(1)}_{\ell} \right]^2
+ \left[ \frac{\zeta^3}{3(\ell - 1)(\ell + 1)} - \frac{\zeta^2}{6(\ell + 1)} \right] \left[ h^{(1)}_{\ell-1} \right]^2
+ \left[ \frac{2\zeta^2}{3(\ell - 1)(\ell + 1)} + \frac{1}{3} \left( h^{(1)}_{\ell}h^{(1)}_{\ell-1} \right) \right.] \tag{5.11}
\]
For \( \ell = 1 \) this expression is singular and cannot be used. From a direct evaluation of the integral for this special case one finds that an exponential integral shows up, as before.

6. Results

The explicit expressions for the basic integrals \( \text{[5.1]} \) that we derived in the previous section can be employed now to determine the corrections to the decay rate. For a perpendicular orientation of the excited atom the correction \( \text{[4.4]} \) to the decay rate is governed by the integrals \( \text{[4.5]} \)–\( \text{[4.6]} \), for which we get upon substitution of the relevant contributions:

\[
J_{\ell,\ell}(\zeta) = \zeta E_1(-2i\zeta) + \left[ \frac{\zeta^3}{6(\ell + 1)} - \frac{\zeta^2}{6(\ell + 1)} \right] \left[ h^{(1)}_{\ell} \right]^2
+ \left[ \frac{\zeta^3}{6(\ell + 1)} - \frac{\zeta^2}{6(\ell + 1)} \right] \left[ h^{(1)}_{\ell-1} \right]^2
+ \left[ \frac{\zeta^4}{3(\ell + 1)} + \zeta^3 \frac{(2\ell^2 + 2\ell - 3)}{3(\ell + 1)} \right] \left[ h^{(1)}_{\ell} \right]^2
- \zeta^3 \sum_{k=1}^\ell \frac{2k + 1}{2(k + 1)} \left[ h^{(1)}_k \right]^2 - \frac{1}{2\zeta^3} \left[ h^{(1)}_0 \right]^2 \tag{6.1}
\]
and
\[ J_{\ell,\perp}^p(\zeta) = \zeta E_1(-2i\zeta) \]
\[ + \left[ \frac{\zeta^5}{6(\ell+1)} - \frac{\zeta^4(2\ell+1)}{3(\ell+1)} \right] h_\ell^{(1)}^2 \]
\[ + \left[ \frac{\zeta^5}{6(\ell+1)} - \frac{2\zeta^4}{3} \right] h_{\ell-1}^{(1)} h_{\ell+1}^{(1)} \]
\[ - \zeta^3 \sum_{k=1}^{\ell} \frac{2k+1}{2(k+1)} \left[ h_k^{(1)} \right]^2 - \frac{1}{2} \zeta^3 \left[ h_0^{(1)} \right]^2, \]
(6.2)
for all \( \ell \geq 1 \). Likewise, the results for a parallel atomic orientation read
\[ J_{\ell,\parallel}^p(\zeta) = 2\zeta E_1(-2i\zeta) \]
\[ + \left[ \frac{\zeta^5}{3(\ell+1)} - \frac{\zeta^4(2\ell^2 + 2\ell - 3)}{3(\ell+1)} \right] h_\ell^{(1)}^2 \]
\[ + \left[ \frac{\zeta^5}{3(\ell+1)} - \frac{2\zeta^4(2\ell^2 + 2\ell - 3)}{3} \right] h_{\ell-1}^{(1)} h_{\ell+1}^{(1)} \]
\[ - \zeta^3 \sum_{k=1}^{\ell} \frac{2k+1}{2(k+1)} \left[ h_k^{(1)} \right]^2 - \zeta^3 \left[ h_0^{(1)} \right]^2, \]
(6.3)
and
\[ J_{\ell,\perp}^p(\zeta) = 2\zeta E_1(-2i\zeta) \]
\[ + \left[ \frac{\zeta^5}{6(\ell+1)} - \frac{\zeta^4(2\ell+1)}{3(\ell+1)} \right] h_\ell^{(1)}^2 \]
\[ + \left[ \frac{\zeta^5}{6(\ell+1)} - \frac{2\zeta^4}{3} \right] h_{\ell-1}^{(1)} h_{\ell+1}^{(1)} \]
\[ - \zeta^3 \sum_{k=1}^{\ell} \frac{2k+1}{2(k+1)} \left[ h_k^{(1)} \right]^2 - \zeta^3 \left[ h_0^{(1)} \right]^2, \]
(6.4)
again for all \( \ell \geq 1 \). As remarked above, the exponential integrals can be expressed in sine and cosine integrals.

After insertion of the expressions (6.1)–(6.2) and the multipole amplitudes (3.5) into (4.4), the average correction to the decay rate for the perpendicular configuration is found in terms of well-known special functions depending on \( \zeta_\alpha, q, \) and \( \varepsilon \). It may be plotted as a function of \( \zeta_\alpha \), for various choices of \( q \) and \( \varepsilon \). To facilitate comparison with our previous results [10] we introduce the decay rate correction function \( f_\perp(\zeta_\alpha, q, \varepsilon) = -16(\Gamma_{\zeta_\alpha})/(3m_0\varepsilon_{0\alpha}) \) with \( \varepsilon_{0\alpha} = 4\pi a^3/3 \) the volume of the spheres.

We shall first consider two special cases that we have treated in [10]: purely scattering spheres and purely absorbing spheres. In the former case we choose the dielectric constant to be real \( (\varepsilon = 1.5) \) and the spherical radius to be finite on the scale of the wavelength \( (q = 0.5) \). For the purely absorbing case with vanishingly small spheres \( (q \rightarrow 0) \), we take the dielectric constant to be complex with the value \( \varepsilon = 1.5 + i0.5 \), as in [10].

Since for small \( q \) the multipole amplitudes \( B_q^\alpha \) and \( B_q^\beta \) behave as \( q^{2\ell+1} \) and \( q^{2\ell+3} \), respectively, only the electric dipole amplitude \( B_1^\alpha = q^2(\pi - 1)/(\varepsilon + 2) \) contributes to (4.4) for the purely absorbing case. In Figs. 1 and 2 the curves for \( f_\perp(\zeta_\alpha) \) are compared to their asymptotic counterparts for large \( \zeta_\alpha \) that follow from (4.8). As can be seen from these figures, the asymptotic curves are quite adequate already for \( \zeta_\alpha \approx 3 \). In the asymptotic regime

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1}
\caption{Decay rate correction function \( f_\perp(\zeta_\alpha) \) (solid line) and its asymptotic form at large distances (dashed line), for a medium with scattering spheres (with \( q = 0.5, \varepsilon(\alpha) = 1.5 \)).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2}
\caption{Decay rate correction function \( f_\perp(\zeta_\alpha) \) (solid line) and its asymptotic form at small and large distances (dashed lines), for a medium with absorbing spheres (with \( q = 0, \varepsilon(\alpha) = 1.5 + i0.5 \)).}
\end{figure}

the results given in [10] are corroborated. (It should be noted that the curves given in [10] erroneously represent \( -f_\perp \) instead of \( f_\perp \).) For small distances the behaviour of the atomic decay rates in the two cases differ considerably. In fact, for the purely scattering case of Fig. 1 the decay rate attains a finite value when \( \zeta_\alpha \) approaches its minimum value \( q \). On the other hand, for the purely absorbing case of Fig. 2 the decay rate correction function is governed by \( J_{\ell,\perp}^p(\zeta_\alpha) \), which according to (6.1) has the asymptotic form \(-1/(4\zeta_\alpha^3)\) for small \( \zeta \). Hence, the decay rate correction function diverges as \(-3/2\) for large \( \zeta_\alpha \) for \( \zeta_\alpha \rightarrow 0 \) in this case.

For a more general situation in which both scattering and absorption take place, we choose \( q = 0.5 \) and \( \varepsilon = 1.5 + i0.5 \), with results presented in Fig. 3. For large \( \zeta_\alpha \) the decay rate falls off like \( \zeta_\alpha^{-1} \), in agreement with (4.8). For small distances the decay rate diverges, as in Fig. 2. In fact, as \( \zeta_\alpha \rightarrow q \), the rate
is found to be proportional to \((\zeta_o - q)^{-1}\). This follows from (4.4), since the series converges increasingly slowly when \(\zeta_o\) approaches \(q\). Indeed, for large \(\ell\) the electric multipole amplitudes \(B^2_\ell\) are given by \(\ell!/(\ell^2(2\ell - 1)!^2)[(\epsilon - 1)/(\epsilon + 1)]q^{2\ell+1}\), while the integral (6.1) gets the form \(-(2\ell - 1)!^2/(2\zeta_o^{2\ell+1})\).

Hence, the electric multipole contribution to the \(\ell\)-th term in the series of (4.4) is \(-\frac{1}{2}[q/(\zeta_o^{2\ell+1})]q^{2\ell+1}\). Since the magnetic multipole contributions turn out to be negligible for large \(\ell\), the asymptotic form of (4.4) for \(\zeta_o\) tending to \(q\) is proportional to \(\sum_{\ell=1}^{\infty} q/(\zeta_o^{2\ell+1}) = q/[2(\zeta_o - q)]\), so that the asymptotic form of \(f_\perp\) reads

\[ f_\perp \approx -\frac{3}{4q^2(\zeta_o - q)} \text{Im} \left[ \frac{\epsilon - 1}{\epsilon + 1} \right] \]  

(6.5)

for \(\zeta_o \to q\).

The physical mechanism for the divergence in the decay rates of Figs. 2 and 3 for small \(\zeta_o\) is the efficient non-radiative energy transfer from the atom to the absorbing spheres that dominates the atomic decay in the near zone. A similar divergent behaviour has been found in a classical treatment of the energy transfer between an excited molecule and a homogeneous absorbing medium filling a halfspace [23]. It should be noted that (6.5) loses its meaning when \(\zeta_o \to q\) becomes so small that the approximations made in deriving it are no longer valid. In particular, perturbation theory in lowest order and the electric-dipole approximation are not adequate to describe the decay for very small distances. Furthermore, the notion of scatterers with a structureless surface gets lost as well in that case.

For the parallel configuration we have likewise evaluated \(f_\parallel(\zeta_o, q, \epsilon) = -16(\Gamma_{a0})/(3\nu_0 \Gamma_{0a})\). The result for the mixed case of both scattering and absorption is given in Fig. 4 for the same choice of the parameters \(q\) and \(\epsilon\) as in Fig. 3. The figure clearly shows that for the parallel configuration the correction to the atomic decay rate goes faster to zero with increasing \(\zeta_o\) than for the perpendicular configuration. This is in accordance with the findings of section 4, where it has been seen that in the asymptotic regime the correction to the atomic decay rate is proportional to the inverse distance in the perpendicular configuration, but to its square in the parallel configuration. As before, the asymptotic expression is adequate from \(\zeta_o \approx 3\) onwards. For small distances, with \(\zeta_o \to q\), the asymptotic form of \(f_\parallel\) is twice that of \(f_\perp\), as follows by comparing the asymptotic forms of (6.1) and (6.3) for large \(\ell\).

In conclusion, we have shown how absorption and scattering processes in a medium may cooperate in modifying the emission rate of an excited atom in its vicinity. The explicit expressions for the decay rate that we have obtained permit a detailed analysis of the behaviour of the emission rate for arbitrary distances between the atom and the medium. As we have seen, the effects of absorption and of scattering are qualitatively different, when the atom approaches the medium.

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