Spontaneous emission and level shifts in absorbing disordered dielectrics and dense atomic gases: A Green's function approach

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Spontaneous emission and Lamb shift of atoms in absorbing dielectrics are discussed. A Green's-function approach is used based on the multipolar interaction Hamiltonian of a collection of atomic dipoles with the quantised radiation field. The rate of decay and level shifts are determined by the retarded Green's-function of the interacting electric displacement field, which is calculated from a Dyson equation describing multiple scattering. The positions of the atomic dipoles forming the dielectrics are assumed to be uncorrelated and a continuum approximation is used. The associated unphysical interactions between different atoms at the same location is eliminated by removing the point-interaction term from the free-space Green's-function (local field correction). For the case of an atom in a purely dispersive medium the spontaneous emission rate is altered by the well-known Lorentz local-field factor. In the presence of absorption a result different from previously suggested expressions is found and nearest-neighbour interactions are shown to be important.

I. INTRODUCTION

The theoretical description and experimental investigation of the interaction of light with dense atomic media regained considerable interest in recent years. Various experiments on level shifts [1,2], intrinsic bistability [3,4] and spontaneous emission [5,6] in dense gases have supported and refined the concept of local fields known for more than a century [7]. Nevertheless some questions in this context are still not answered satisfactory even on a fundamental level. In the present paper I want to discuss one of these questions, namely the effect of an *absorbing* dielectric on spontaneous emission and level shifts of an embedded atom using a Green's-function approach.

The interaction of light with dilute gases is usually well described in terms of macroscopic classical variables such as electric field and polarisation. In the macroscopic approach the polarisation is given by the expectation value of the single-atom dipole moment multiplied by the density of atoms [8]. Apart from the coupling to the common classical radiation field, the atoms are assumed distinguishable and independent. This means quantum-statistical correlations are neglected, which is a very good approximation as long as the temperatures are not too small. It is also implicitly assumed that vacuum fluctuations of the field affect the atoms only individually and that the atom positions are independent of each other. The latter assumptions are however no longer valid in dense samples.

If the resonant absorption length of some atomic transition becomes comparable to the medium dimension d, i.e. for $N\lambda^2 d \sim 1$, N being the number density and λ the resonant wavelength, reabsorption and multiple scattering of spontaneous photons and associated effects like radiation trapping [9] or, if atomic excitation is present, amplified spontaneous emission need to be taken into account. If the atomic density is further increased, such that $N\lambda^3 \sim 1$, one can no longer disregard the fact that the independent-atom approximation allows for an unphysical interaction of different atoms at the *same* position and Lorentz-Lorenz local field corrections are needed [7].

The modification of the rate of spontaneous emission Γ by the local environment was first noted by Purcell [11]. Alterations of this rate have been demonstrated experimentally near dielectric interfaces [12], in quantum-well structures [13] and in cavities [14]. Based on an analysis of the density of radiation states Nienhuis and Alkemade predicted for an atom embedded in a homogeneous transparent dielectric with refractive index n [15]:

$$\Gamma = \Gamma_0 n \tag{1}$$

where Γ_0 is the free-space decay rate.

$$\Gamma_0 = \frac{\wp^2 \omega_{ab}^3}{3\pi\hbar\epsilon_0 c^3}, \tag{2}$$

$\wp$ being the electric dipole moment of the transition with frequency ω_{ab}. The alteration of spontaneous emission by the index of refraction leads to interesting potential applications as the suppression or enhancement of decay in photonic band-gap materials [16]. The approach of Ref. [15] did neither take into account local-field corrections nor absorption however.

There has been a considerable amount of theoretical work on local-field corrections to spontaneous emission of an atom in *lossless* homogeneous dielectrics. Essentially all approaches assume a small cavity around the radiating atom and the theoretical predictions depend substantially on the details of this local-cavity model. Approaches based on Lorentz's "virtual" cavity [17,18] lead to

$$\Gamma_{\rm Lor} = \Gamma_0\, n\, \left(\frac{n^2+2}{3}\right)^2, \tag{3}$$

while those based on a real empty cavity [19] predict

$$\Gamma_{\rm emp} = \Gamma_0\, n\, \left(\frac{3n^2}{2n^2+1}\right)^2. \tag{4}$$

For pure systems or impurities in disordered non-absorbing dielectrics Eq.(3) is believed to be correct. On the other hand recent experiments with Eu^{3+} ions in organic ligand cages verified the real-cavity expression Eq.(4) [5,6]. An explanation for the different results was very recently given by de Vries and Lagendijk [20]. Applying a rigorous microscopic scattering theory for impurities in non-absorbing dielectric cubic crystals, they showed that the local environment determines whether Eq.(3) or (4) should be used. For a substitutional impurity the empty-cavity result applies, while for an interstitial impurity the virtual-cavity formula is correct. The latter also supports the believe that Eq.(3) is the correct one for disordered systems like gases.

While the effect of a transparent dielectric on spontaneous emission is rather well studied, this it not the case for *absorbing* media. A first step in this direction was made by Barnett, Huttner and Loudon [21]. Based on a discussion of the retarded Green's-function in an absorbing bulk dielectric they showed, that the index of refraction in (1) is to be replaced by the real part n' of the complex refractive index $n = n' + in''$. They also argued that the square of the Lorentz-local field factor in (3) should be replaced by the absolute square, leading to

$$\Gamma = \Gamma_0\, n'(\omega_{ab}) \left|\frac{n^2(\omega_{ab})+2}{3}\right|^2. \tag{5}$$

In order to derive this equation Barnett et al. postulated in [22] an operator equivalent of the Lorentz-Lorenz relation between the Maxwell and local field. This assumption has however some conceptual problems. As pointed out very recently by Scheel et al. [23], an operator Lorentz-Lorenz relation cannot hold, since both quantities, the Maxwell field and the local field have to fulfil the same commutation relations.

In a recent paper we have developed an approach that takes into account local-field corrections as well as multiple scattering and reabsorption of spontaneous photons in modified single-atom Bloch equations [10]. The modified Bloch equations provide a way of including dense-medium effects in a macroscopic approach. In the present paper expressions for the spontaneous emission rate and Lamb-shift of an atom in a dense *absorbing* dielectric or a gas of identical atoms are derived following the approach of [10]. The starting point is the multipolar-coupling Hamiltonian in dipole approximation. The retarded Green's-function of the electric displacement field, which determines decay rate and Lamb shift, is calculated from a Dyson equation in self-consistent Hartree approximation. As the atom positions are assumed to be independent from each other, local-field corrections are needed to remove unphysical interactions between atoms at zero distance. This is done in the present approach by an appropriate modification of the free-space Green's-functions rather than by introducing a cavity. The rate of spontaneous emission derived coincides with the virtual-cavity result (3) for a transparent dielectric, but differs from Eq.(5) in the case of absorption. It will be shown that in the presence of absorption near-field interactions with neighbouring atoms become very important, whose correct description requires however a fully microscopic approach.

II. RADIATIVE INTERACTIONS IN DENSE ATOMIC MEDIA

The present analysis is based on a description of the atom-field interaction in the dipole approximation using the multipolar Hamiltonian in the radiation gauge [24]

$$\hat{H}_{\rm int} = -\frac{1}{\epsilon_0}\sum_j \hat{\vec{d}}_j \cdot \hat{\vec{D}}(\vec{r}_j). \tag{6}$$

Here $\hat{d}_j$ is the dipole operator of an atom at position $\vec{r}_j$. $\hat{D}$ is the operator of the electric displacement with $\nabla\cdot\hat{\vec{D}} = 0$.

It was shown in [10] that the effects of radiative atom-atom interactions in a dense medium can be described in Markov approximation with a nonlinear density-matrix equation

$$\begin{aligned}
\dot\rho = &-\frac{i}{\hbar}\Big[H_0,\rho\Big] + i\frac{\wp_\mu}{\hbar}\Big[\sigma_\mu\mathcal{E}_{L\mu}^- + \sigma_\mu^\dagger\mathcal{E}_{L\mu}^+,\rho\Big]\\
&-ih_{\mu\nu}\Big[\sigma_\nu^\dagger\sigma_\mu,\rho\Big] - ih_{\mu\nu}^c\Big[\Big[\sigma_\nu^\dagger,\sigma_\mu\Big],\rho\Big]\\
&-\frac{\Gamma_{\mu\nu}}{2}\Big\{\sigma_\nu^\dagger\sigma_\mu\rho + \rho\sigma_\nu^\dagger\sigma_\mu - 2\sigma_\mu\rho\sigma_\nu^\dagger\Big\}\\
&-\frac{\Gamma_{\mu\nu}^c}{2}\Big\{\Big[\sigma_\mu,\big[\sigma_\nu^\dagger,\rho\big]\Big] + \Big[\sigma_\nu^\dagger,\big[\sigma_\mu,\rho\big]\Big]\Big\}.
\end{aligned} \tag{7}$$

Here $\wp_\mu$ is the dipole matrix element for a polarisation direction $\vec{e}_\mu$ and $\sigma_\mu, \sigma_\mu^\dagger$ are the corresponding atomic lowering and razing operators. The first term describes the free atomic evolution and the second the interaction with some local classical field $\mathcal{E}_L$. $h_{\mu\nu}$ and $\Gamma_{\mu\nu}$ are matrices, whose eigenvalues yield Lamb shifts of excited states and spontaneous emission rates. $\Gamma_{\mu\nu}^c$ and $h_{\mu\nu}^c$ describe collective relaxation rates and light-shifts due to the incoherent background radiation generated by absorption and reemission of spontaneous photons (radiation trapping).

It should be noted that the incoherent background radiation causes a decay as well as an incoherent excitation with equal rate Γ^c. Thus Γ^c, which is proportional to the excitation of the host medium [10], describes *induced* mixing processes, while Γ can be interpreted as the rate of *spontaneous* decay. Similarly h^c describes a light-shift, which for a two level system is equal in strength and opposite in sign for the ground and excited state. It is also proportional to the excitation of the host medium and can thus be interpreted as *induced* light shift. In contrast h is a frequency shift of an excited state only and does not require excitation of the host medium.

The matrices $\Gamma_{\mu\nu}$ and $h_{\mu\nu}$ are given by [10]

$$\Gamma_{\mu\nu} = 2\frac{\wp_\mu \wp_\nu}{\hbar^2}\,\mathrm{Re}\Big[\mathrm{D}_{\mu\nu}(0,\omega_{ab})\Big], \tag{8}$$

$$h_{\mu\nu} = \frac{\wp_\mu \wp_\nu}{\hbar^2}\,\mathrm{Im}\Big[\mathrm{D}_{\mu\nu}(0,\omega_{ab})\Big], \tag{9}$$

where $\mathrm{D}_{\mu\nu}(\vec{x},\omega) \equiv \int_{-\infty}^{\infty} \mathrm{d}\tau\, D_{\mu\nu}(\vec{x},\tau)\mathrm{e}^{i\omega\tau}$ is the Fourier-transform of the retarded Green's-function (GF) of the electric displacement field defined here as

$$\mathrm{D}_{\mu\nu}(\vec{x},\tau) = \theta(\tau)\big\langle 0\big|\Big[\hat{D}_\mu(\vec{r}_1,t_1),\hat{D}_\nu(\vec{r}_2,t_2)\Big]\big|0\big\rangle\,\epsilon_0^{-2}, \tag{10}$$

with $\vec{x} = \vec{r}_1 - \vec{r}_2$ and $\tau = t_1 - t_2$. In the case of randomly oriented two-level atoms, one can replace $\wp_\mu \to \wp$ and perform an orientation average yielding a single decay rate Γ and a single excited-state level shift h.

The dense atomic medium affects the spontaneous emission of a single probe atom due to multiple scattering of virtual photons. The scattering process can formally be described by a Dyson equation for the exact retarded GF

$$\mathbf{D}(1,2) = \mathbf{D}^0(1,2) - \iint d3\, d4\, \mathbf{D}^0(1,3)\,\mathbf{\Pi}(3,4)\,\mathbf{D}(4,2). \tag{11}$$

Here the integration is over t from $-\infty$ to $+\infty$ and the whole sample volume. $\mathbf{D}^0$ is the (dyadic) GF in free space and $\mathbf{\Pi}$ is a formal (dyadic) self-energy. As shown in [10], the self-energy can be described for randomly oriented two-level atoms in self-consistent Hartree approximation by

$$\mathbf{\Pi}(1,2) = \sum_j \frac{2}{3}\frac{\wp^2}{\hbar^2}\theta(t_1-t_2)\Big\langle\big[\sigma_j^\dagger(t_1),\sigma_j(t_2)\big]\Big\rangle \times\delta(\vec{r}_1-\vec{r}_j)\delta(\vec{r}_2-\vec{r}_j)\,\mathbf{1}. \tag{12}$$

$\mathbf{1}$ is unity matrix and $\sigma = |b\rangle\langle a|$ is the atomic spin-flip operator from the excited state $|a\rangle$ to the lower state $|b\rangle$ in the Heisenberg picture, i.e. it contains all interactions. The factor $2/3$ results from an orientation average.

We now make a continuum approximation and assume a homogeneous medium, such that

$$\mathbf{\Pi}(1,2) \longrightarrow p(t_1,t_2)\delta(\vec{r}_1-\vec{r}_2)\,\mathbf{1}, \tag{13}$$

where

$$p(t_1,t_2) = \frac{2}{3}\frac{\wp^2}{\hbar^2}N\theta(t_1-t_2)\overline{\Big\langle\big[\sigma^\dagger(t_1),\sigma(t_2)\big]\Big\rangle}. \tag{14}$$

The over-bar denotes an average over some possible inhomogeneous distribution and N is the number density of atoms.

With the above made approximations, the Dyson equation (11) contains also scattering processes between atoms at the same position. In a continuum approximation the probability of two point dipoles being at the same position is of measure zero. This nevertheless leads to a non-vanishing contribution, since the dipole-dipole interaction has a δ-type point interaction. This unphysical contribution needs to be removed by a local-field corrections, which will be discussed in the following section.

III. LOCAL-FIELD CORRECTION OF FREE-SPACE GREEN'S-FUNCTION AND LORENTZ-LORENZ RELATION

The retarded Green's-function in free space $\mathrm{D}^0_{\mu\nu}(1,2) = \theta(t_1-t_2)\langle 0\big|\big[\hat{D}^0_\mu(1),\hat{D}^0_\nu(2)\big]\big|0\rangle\epsilon_0^{-2}$, where $1;2;\ldots$ stand for $\vec{r}_1,t_1;\vec{r}_2,t_2;\ldots$ etc., is a solution of the homogeneous Maxwell equation with δ-like source term

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + \nabla\times\nabla\times\right)\mathbf{D}^0(1,2) = -\frac{i\hbar}{\epsilon_0}\frac{\omega^2}{c^2}\delta(\vec{r}_1-\vec{r}_2)\delta(t_1-t_2)\,\mathbf{1}. \tag{15}$$

$\mathbf{D}^0$ has a particularly simple form in reciprocal space [25]

$$\mathbf{D}^0(\vec{q},\omega^+) = \frac{i\hbar}{\epsilon_0}\frac{k^2}{(k^2+i0)\,\mathbf{1} - q^2\mathbf{\Delta}_q} \tag{16}$$

$$= \frac{i\hbar}{\epsilon_0}\left[\frac{k^2}{k^2-q^2+i0}\mathbf{\Delta}_q + \frac{\vec{q}\circ\vec{q}}{q^2}\right], \tag{17}$$

where $k = \omega/c$, and $\mathbf{\Delta}_q = \mathbf{1} - \frac{\vec{q}\circ\vec{q}}{q^2}$. It may be worthwhile noting, that $\mathbf{D}^0$ is not transverse in $\vec{q}$-space, although $\nabla_1\cdot\mathbf{D}^0(1,2) \equiv 0$. The corresponding function in coordinate space reads [25]

$$\mathbf{D}^0(\vec{x},\omega^+) = -\frac{i\hbar\omega^2}{\epsilon_0 c^2}\frac{\mathrm{e}^{ik^+x}}{4\pi x}\left[P\,(ikx)\;\mathbf{1} + Q\,(ikx)\,\frac{\vec{x}\circ\vec{x}}{x^2}\right] + \frac{i\hbar}{3\epsilon_0}\delta(\vec{x})\,\mathbf{1}. \tag{18}$$

Here $x = |\vec{x}|$ and

$$P(z) = 1 - \frac{1}{z} + \frac{1}{z^2}, \qquad Q(z) = -1 + \frac{3}{z} - \frac{3}{z^2}. \tag{19}$$

One recognises from Eq.(18) that the retarded GF of the dipole-dipole interaction contains a δ-type point contribution. In order to eliminate the unphysical interactions between different atoms at the same position, one has to remove this term from the GFs in the scattering part of the Dyson equation (11).

$$\mathbf{D}^0(\vec{x},\omega^+) \longrightarrow \mathbf{F}^0(\vec{x},\omega^+) = \mathbf{D}^0(\vec{x},\omega^+) - \frac{i\hbar}{3\epsilon_0}\delta(\vec{x})\mathbf{1}. \quad (20)$$

With this local-field corrections we obtain a modified Dyson equation (in reciprocal space)

$$\mathbf{D} = \mathbf{D}^0 - \mathbf{F}^0 p \mathbf{F}^0 + \mathbf{F}^0 p \mathbf{F}^0 p \mathbf{F}^0 - + \cdots, \quad (21)$$

and introducing $\mathbf{F}(\vec{q},\omega) \equiv \mathbf{D}(\vec{q},\omega) - i\hbar/3\epsilon_0\,\mathbf{1}$ we arrive at

$$\mathbf{F}(\vec{q},\omega^+) = \mathbf{F}^0(\vec{q},\omega^+) - \mathbf{F}^0(\vec{q},\omega^+)\,p(\omega^+)\,\mathbf{F}(\vec{q},\omega^+). \quad (22)$$

In reciprocal space one finds

$$\mathbf{F}^0(\vec{q},\omega^+) = \mathbf{D}^0(\vec{q},\omega^+) - \frac{i\hbar}{3\epsilon_0}\mathbf{1}, \quad (23)$$

$$= -\frac{i\hbar}{\epsilon_0}\left[\frac{\left(\frac{1}{3}q^2 + \frac{2}{3}k^2\right)\mathbf{1} - \vec{q}\circ\vec{q}}{q^2 - k^2 - i\epsilon}\right]. \quad (24)$$

Eq.(22) can easily be solved to yield

$$\mathbf{F}(\vec{q},\omega^+) = -\frac{i\hbar}{\epsilon_0}\left[\frac{\left(\frac{1}{3}q^2 + \frac{2}{3}k^2\right)\left(1 + \frac{2}{3}N\alpha(\omega)\right)\mathbf{1} - \vec{q}\circ\vec{q}}{q^2 - k^2 - N\alpha(\omega)\left(\frac{1}{3}q^2 + \frac{2}{3}k^2\right) - i0}\right] \times \frac{1}{1 + \frac{2}{3}N\alpha(\omega)} \quad (25)$$

where we have introduced the dynamic polarisability of the atoms

$$N\alpha(\omega) \equiv \frac{i\hbar}{\epsilon_0}p(\omega). \quad (26)$$

The poles $\pm q_0$ of Eq.(25) determine the (in general non-linear) complex dielectric function

$$\varepsilon(\omega) \equiv \frac{q_0^2}{k^2} = 1 + \frac{N\alpha(\omega)}{1 - \frac{1}{3}N\alpha(\omega)}. \quad (27)$$

This is the well-known Lorentz-Lorenz relation between the microscopic polarisability α and the complex dielectric function $\varepsilon(\omega)$. Thus we have shown that the local-field correction of the free-space Green's-function (20) is exactly the one that reproduces the well-known Lorentz-Lorenz relation.

IV. MODIFICATION OF SPONTANEOUS EMISSION AND LAMB-SHIFT

Eq.(25) can be transformed back into coordinate space using $\widetilde{\mathrm{F}}(\vec{x},\omega^+) = (2\pi)^{-3}\int \mathrm{d}^3\vec{q}\,\widetilde{\mathrm{F}}(\vec{q},\omega^+)\,\mathrm{e}^{-i\vec{q}\vec{x}}$. The Fourier-transform of the projector $(\vec{q}\circ\vec{q})$ yields spherical Bessel functions [25]. For the present purpose we however need only the orientation-averaged quantity

$$\mathrm{F}(\vec{q},\omega^+) = -\frac{2i\hbar}{3\epsilon_0}\left[\frac{\frac{1}{3}q^2 N\alpha(\omega) + k^2\left(1 + \frac{2}{3}N\alpha(\omega)\right)}{q^2 - k^2 - N\alpha(\omega)\left(\frac{1}{3}q^2 + \frac{2}{3}k^2\right) - i0}\right] \times \frac{1}{1 + \frac{2}{3}N\alpha(\omega)}. \quad (28)$$

One recognises that the Fourier-transform of $\mathrm{F}(\vec{q},\omega^+)$ diverges for $x \to 0$, which is due to the large-q behaviour of the GF. In order to remove these singularities one can modify the GF by introducing a regularisation. Physically the singular behaviour at $x \to 0$ is due to the fact that atoms very close to the atom under consideration can have a large effect on spontaneous emission and level shifts. One cannot expect the continuum approximation used here to yield accurate results on length scales comparable to the mean atom distance. Here rather a fully microscopic description of very close atoms including their motion (collisions) is needed. This is however beyond the scope of the present paper and we therefore restrict the analysis to a regularisation of the Green's-function. There is no unique regularisation procedure, and we here just choose a convenient one

$$\mathrm{F}(\vec{q},\omega^+) \longrightarrow \widetilde{\mathrm{F}}(\vec{q},\omega^+) = \mathrm{F}(\vec{q},\omega^+)\frac{\Lambda^4}{q^4 + \Lambda^4}. \quad (29)$$

With this we find in the limit $\Lambda \gg |q_0|$

$$\widetilde{\mathrm{F}}(\vec{x}=0,\omega^+) = \frac{\hbar\omega^3}{6\pi\epsilon_0 c^3}\sqrt{\varepsilon(\omega)}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2 - \frac{i\hbar\omega^3}{6\pi\epsilon_0 c^3}\left[\frac{1}{R}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2 + \frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3}\right)(\varepsilon(\omega)-1)\right], \quad (30)$$

where $R = k/(\sqrt{2}\Lambda)$. It is important to note, that $\widetilde{\mathrm{F}}$ is exactly causal, if $\varepsilon(\omega)$ fulfils the Krames-Kronig relations. This would not have been the case if as according to the result of Barnett et al. [21,22] the absolute square $|(\varepsilon+2)/3|^2$ would be present instead of $((\varepsilon+2)/3)^2$.

With this result we find for the decay rate and excited state Lamb-shift

$$\Gamma = \Gamma_0\,\mathrm{Re}\left[\sqrt{\varepsilon(\omega)}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2\right] + \Gamma_0\,\mathrm{Im}\left[\frac{1}{R}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2 + \frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3}\right)(\varepsilon(\omega)-1)\right], \quad (31)$$

$$h = \frac{\Gamma_0}{2}\,\mathrm{Im}\left[\sqrt{\varepsilon(\omega)}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2\right] - \frac{\Gamma_0}{2}\,\mathrm{Re}\left[\frac{1}{R}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2 + \frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3}\right)(\varepsilon(\omega)-1)\right]. \quad (32)$$

For an atom in a purely *dispersive* disordered medium, i.e. for $\varepsilon'' \equiv 0$, the second term in Eq.(31) for the spontaneous decay rate vanishes identically and we are left with the "virtual" cavity result Eq.(3). Likewise there are no contributions from the first term in (32) to the Lamb shift in this case.

In the presence of absorption, that is if the probe-atom transition frequency comes closer to a resonance of the surrounding material (as it would naturally be the case for a collection of identical atoms) Γ is different from the result obtained in [21,22,28]. In this case there are also non-vanishing terms that contain the regularisation parameter R^{-1} and R^{-3}. These terms must be interpreted as contributions due to resonant energy transfer with nearest neighbours, a process which cannot accurately be described in the continuum approach used here.

As the Lamb shift is concerned, Eq.(32) shows that in a purely dispersive medium, that is far away from any resonances only nearest-neighbour interactions matter. This is intuitively clear since in this case the transition frequency is only affected by collisions. Only in the presence of absorption there is also a bulk-contribution to the Lamb shift as described by the first term in (32).

For a dense gas of identical atoms or of atoms of the same kind but with some inhomogeneous broadening, Eqs.(31) and (32) are only implicit, since the complex polarisability ε depends on the decay rate and level shift. Hence a self-consistent determination of Γ and h is necessary. If the density of atoms is much less than one per cubic wavelength one can consider an expansion of Γ and h in powers of the atomic density N. Defining $\alpha = \alpha' + i\alpha''$ one finds with Eq.(27) for the bulk contributions

$$\Gamma = \Gamma_0 \left[1 + \frac{7}{6}\alpha' N + \frac{17}{24}\left(\alpha'^2 - \alpha''^2\right) N^2 + \mathcal{O}(N^3) \right], \quad (33)$$

$$h = \frac{\Gamma_0}{2} \left[\frac{7}{6}\alpha'' N + \frac{17}{12}\alpha'\alpha'' N^2 + \mathcal{O}(N^3) \right]. \quad (34)$$

In the case of radiatively broadened two-level atoms, the real part of atomic polarisability vanishes at resonance, i.e. $\alpha' = 0$. Thus in lowest order of the density there is only a contribution to the excited state frequency proportional to the population difference between excited and ground state. For an inverted population the transition frequency is red-shifted, for balanced population the level shift vanishes and for more atoms in the lower state the transition frequency is blue shifted. As a result spontaneously emitted radiation from an initially inverted system will have a chirp very similar to the chirp in Dicke-superradiance [26]. It should also be mentioned that the shift of the transition frequency discussed here is physically different from the familiar Lorentz-Lorenz shift. The LL-shift is due to the dispersion of the index of refraction at an atomic resonance and is thus in contrast to the absorption α'' independent on Doppler-broadening [27].

V. SUMMARY

In the present paper we have discussed the rate of spontaneous emission and the excited-state level shift of a two-level type probe atom inside a homogeneous, disordered absorbing dielectric. The dielectric was modelled by a collection of atomic point dipoles, which also includes the case of a dense gas of identical atoms. The multiple scattering of photons between the atoms (dipole-dipole interaction) was described by a Dyson integral equation for the exact retarded Green's-function of the electric displacement field in self-consistent Hartree approximation. The atoms were assumed distinguishable with random independent positions. The latter assumption made a continuum approximation possible and the Dyson equation could be solved analytically. In order to exclude unphysical dipole-dipole interactions of different atoms at the same position arising in the continuum approximation with independent atomic positions, a local-field correction of the free-space retarded Green's-function was introduced. This lead to the well-known Lorentz-Lorenz relation between the complex dielectric function $\varepsilon(\omega)$ and the nonlinear atomic polarisability $\alpha(\omega)$. The expression for the spontaneous-decay rate found by this method agrees with the virtual cavity result [17] in the absence of absorption. This is an expected result for atoms in disordered dielectrics [20]. It was shown that the excited-state Lamb shift is in this case only affected by nearest-neighbour interactions, which could not be treated accurately within the present approach however. In the presence of absorption the spontaneous-emission rate differs from the results obtained in [21,22,28] in two ways. First there are important nearest-neighbour contributions, which were absent in the models of [21,22]. Secondly the bulk-contribution is different form Refs. [21,22,28], since causality of the exact retarded GF requires the Lorentz-field factor to enter as square and not as absolute square. It is interesting to note, that apart from a small difference in the R^{-3} term, the decay rate derived here is identical to one very recently obtained by Scheel and Welsch [29] on the basis of a completely different approach, namely a quantisation of the electromagnetic field in a linear dielectric.

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