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

Michael Fleischhauer

Sektion Physik, Ludwig-Maximilians Universität München, D-80333 München, Germany

Spontaneous emission and Lamb shift of atoms in absorbing dielectrics and dense atomic gases are discussed using a microscopic Green's-function approach. Uncorrelated and random atomic positions are assumed, and the associated unphysical interactions between different atoms at the same location are eliminated (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. When the mean distance between atoms becomes less than the resonance wavelength, results different from previously suggested expressions are found. In particular, it is shown that nearest-neighbor interactions become important. The results suggest that, for large densities, absorbing disordered dielectrics cannot accurately be described by a macroscopic approach that neglects correlations between atomic positions. [S1050-2947(99)08309-2]

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 polarization. In the macroscopic approach the polarization 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 such as 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] took into account neither 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

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

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

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

For pure systems or impurities in disordered nonabsorbing 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 nonabsorbing dielectric cubic crystals, they showed that the local environment determines whether Eq. (3) or Eq. (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 belief that Eq. (3) is the correct one for disordered systems such as 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 Eq. (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 Eq. (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 fulfill 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 the dipole approximation. The retarded Green's function of the electric displacement field, which determines the decay rate and Lamb shift, is calculated from a Dyson equation in the 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 neighboring atoms become very important, the correct description of which 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],

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

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 $\vec{\nabla} \cdot \hat{\vec{D}} = \mathbf{0}$.

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

$$\dot{\rho} = -\frac{i}{\hbar}[H_0, \rho] + i\frac{\wp_\mu}{\hbar}[\sigma_\mu \mathcal{E}_{L\mu}^- + \sigma_\mu^\dagger \mathcal{E}_{L\mu}^+, \rho]$$

$$- ih_{\mu\nu}[\sigma_\nu^\dagger \sigma_\mu, \rho] - ih_{\mu\nu}^c[[\sigma_\nu^\dagger, \sigma_\mu], \rho]$$

$$- \frac{\Gamma_{\mu\nu}}{2}\{\sigma_\nu^\dagger \sigma_\mu \rho + \rho \sigma_\nu^\dagger \sigma_\mu - 2\sigma_\mu \rho \sigma_\nu^\dagger\}$$

$$- \frac{\Gamma_{\mu\nu}^c}{2}\{[\sigma_\mu, [\sigma_\nu^\dagger, \rho]] + [\sigma_\nu^\dagger, [\sigma_\mu, \rho]]\}. \tag{7}$$

Here $\wp_\mu$ is the dipole matrix element for a polarization 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 an *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]

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

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

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

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

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 $\wp_\mu\rightarrow\wp$ 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,

$$\mathbf{D}(1,2)=\mathbf{D}^0(1,2)-\int\int d3d4\mathbf{D}^0(1,3)\mathbf{\Pi}(3,4)\mathbf{D}(4,2). \tag{11}$$

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 the self-consistent Hartree approximation by

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

$\mathbf{1}$ is the 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

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

where

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

The overbar 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 nonvanishing 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 THE FREE-SPACE GREEN'S-FUNCTION AND LORENTZ-LORENZ RELATION

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

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

$\mathbf{D}^0$ 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 $\mathbf{\Delta}_q=\mathbf{1}-\vec{q}\circ\vec{q}/q^2$. Note that $\mathbf{D}^0$ is not transverse although $\mathbf{\nabla}_1\cdot\mathbf{D}^0(1,2)\equiv 0$ for $\vec{r}_1\neq\vec{r}_2$. The corresponding function in coordinate space reads [25]

$$\mathbf{D}^0(\vec{x},\omega^+)=-\frac{i\hbar\omega^2}{\epsilon_0c^2}\frac{e^{ik^+x}}{4\pi x}\left[P(ikx)\mathbf{1}+Q(ikx)\frac{\vec{x}\circ\vec{x}}{x^2}\right]$$
$$+\frac{i\hbar}{3\epsilon_0}\delta(\vec{x})\mathbf{1}. \tag{18}$$

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

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

One recognizes 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),

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

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

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

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

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

In reciprocal space one finds

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

$$=-\frac{i\hbar}{\epsilon_0}\left[\frac{\left(\frac{1}{3}q^2+\frac{2}{3}k^2\right)\mathbf{1}-\vec{q}\circ\vec{q}}{q^2-k^2-i0}\right]. \tag{24}$$

Equation (22) can easily be solved to yield

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

$$\times\frac{1}{1+\frac{2}{3}N\alpha(\omega)}, \tag{25}$$

where we have introduced the dynamic polarizability of the atoms

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

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

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

This is the well-known Lorentz-Lorenz relation between the microscopic polarizability 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

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

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

$$\times\frac{1}{1+\frac{2}{3}N\alpha(\omega)}. \tag{28}$$

One recognizes that the Fourier transform of $F(\vec{q},\omega^+)$ diverges for $x\rightarrow 0$, which is due to the large-q behavior of the GF. In order to remove these singularities, one can modify the GF by introducing a regularization. Physically

the singular behavior at $x\rightarrow 0$ 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 regularization of the Green's function. There is no unique regularization procedure, and here we just choose a convenient one,

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

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

$$\widetilde{F}(\vec{x}=0,\omega^+)=\frac{\hbar\omega^3}{6\pi\epsilon_0 c^3}\sqrt{\varepsilon(\omega)}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2$$

$$-\frac{i\hbar\omega^3}{6\pi\epsilon_0 c^3}\left[\frac{1}{R}\left(\frac{\varepsilon(\omega)+2}{3}\right)^2\right.$$

$$\left.+\frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3\varepsilon(\omega)}\right)[\varepsilon(\omega)-1]\right], \tag{30}$$

where $R=k/(\sqrt{2}\Lambda)$. It is important to note that $\widetilde{F}$ is exactly causal, if $\varepsilon(\omega)$ fulfills the Krames-Kronig relations. This would not have been the case if, 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

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

$$\left.+\frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3\varepsilon(\omega)}\right)[\varepsilon(\omega)-1]\right], \tag{31}$$

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

$$\left.+\frac{1}{R^3}\frac{2}{3}\left(\frac{\varepsilon(\omega)+2}{3\varepsilon(\omega)}\right)[\varepsilon(\omega)-1]\right]. \tag{32}$$

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 Eq. (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 nonvanishing terms that contain the regularization parameter R^{-1} and

R^{-3}. These terms must be interpreted as contributions due to resonant energy transfer with nearest neighbors. This process cannot accurately be described in the present approach, which ignores correlations between atom positions generated by hard-core repulsion and dipole forces associated with the level shifts.

As the Lamb shift is concerned, Eq. (32) shows that in a purely dispersive medium, that is, far away from any resonances, only nearest-neighbor interactions matter. This is intuitively clear since in this case the transition frequency is only affected by dipole-dipole interactions of close neighbors. Only in the presence of absorption is there also a bulk contribution to the Lamb shift as described by the first term in Eq. (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 polarizability ε depends on the decay rate and level shift as well as short-range correlations of atomic positions. Hence a self-consistent determination of Γ and h is necessary. If the density is much less than one atom 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

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

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

In the case of radiatively broadened two-level atoms, the real part of atomic polarizability 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 the excited and the ground state. For an inverted population the transition frequency is redshifted, for balanced population the level shift vanishes, and for more atoms in the lower state the transition frequency is blueshifted. 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 spon-

taneous emission and the excited-state level shift of a two-level type probe atom inside a homogeneous, disordered absorbing dielectric. The dielectric was modeled 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 led to the well-known Lorentz-Lorenz relation between the complex dielectric function $\varepsilon(\omega)$ and the nonlinear atomic polarizability $\alpha(\omega)$. 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-neighbor 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-neighbor contributions, which were absent in the models of [21,22]. Second, 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 a square and not as an absolute square. It is interesting to note that apart from an overall numerical prefactor (which depends on the details of the regularization procedure), 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 quantization of the electromagnetic field in a linear dielectric. The presence of nearest-neighbor contributions suggests that a macroscopic description that ignores correlations between atomic positions is no longer valid when the mean distance between atoms becomes comparable to the resonance wavelength of a dipole transition.

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