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Multiple-scattering approach to interatomic interactions and superradiance in inhomogeneous dielectrics

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The dynamics of a collection of resonant atoms embedded inside an inhomogeneous nondispersive and lossless dielectric is described with a dipole Hamiltonian that is based on a canonical quantization theory. The dielectric is described macroscopically by a position-dependent dielectric function and the atoms as microscopic harmonic oscillators. We identify and discuss the role of several types of Green tensors that describe the spatio-temporal propagation of field operators. After integrating out the atomic degrees of freedom, a multiple-scattering formalism emerges in which an exact Lippmann-Schwinger equation for the electric field operator plays a central role. The equation describes atoms as point sources and point scatterers for light. First, single-atom properties are calculated such as position-dependent spontaneous-emission rates as well as differential cross sections for elastic scattering and for resonance fluorescence. Secondly, multiatom processes are studied. It is shown that the medium modifies both the resonant and the static parts of the dipole-dipole interactions. These interatomic interactions may cause the atoms to scatter and emit light cooperatively. Unlike in free space, differences in position-dependent emission rates and radiative line shifts influence cooperative decay in the dielectric. As a generic example, it is shown that near a partially reflecting plane there is a sharp transition from two-atom superradiance to single-atom emission as the atomic positions are varied.

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I. INTRODUCTION

The spontaneous-emission rate of an atom depends on its dielectric environment [1,2] and in particular on the precise position of the atom if the medium is inhomogeneous [3–8]. Spontaneous emission can only be understood quantum mechanically, but the classical Green function determines the emission rate. In particular, the emission rate is proportional to the imaginary part of the Green tensor of the medium at the atomic position. The dipole-angle average of the emission rate is also known as the local optical density of states [9].

In order to study the influence of the medium on much more than just single-atom spontaneous-emission rates, in this paper a rather general multiple-scattering theory is set up. It is based on "macroscopic quantization" theories of the electromagnetic field in inhomogeneous lossless dielectrics, see for example [5,10–12]. Microscopic treatments of (macroscopically homogeneous) dielectrics in quantum electrodynamics can be found in [13,14] but will not be used here. An important result in the macroscopic theories is that photons can be defined as the elementary excitations of the true modes of the dielectric. Corresponding mode functions are the (classical) harmonic solutions of the wave equation.

Emission rates of an atom not only change due to the nonresonant dielectric environment, but also due to the presence of other atoms with the same transition frequency. As is known since the pioneering work by Dicke [15], resonant atoms in each other's neighborhood decay cooperatively. Depending on the many-atom state, the atoms decay faster than a single atom up till twice the single-atom rate (superradiance) or decay slower or not at all (subradiance). Lifetime changes of individual atom pairs as a function of their distance were measured only recently [16]; for two Ba^+ ions that emit at a wavelength of 493 nm and for well-defined separations $|\mathbf{R}|$ around 1.5 μm, subradiant and superradiant lifetime effects of less than ±2% were observed.

Superradiance occurs for the so-called Dicke states that have a zero expectation value of the total dipole moment [15], but also for atomic product states with a nonzero dipole moment [17,18]. Superradiance also occurs for classical dipoles. It is a general phenomenon also exhibited in acoustics by nearby identical tuning forks, or by strings in a piano [18]. These systems have in common that the atoms (or oscillators) interact with a field that is influenced by the radiation reactions of all nearby atoms together.

Cooperative effects of resonant atoms will be influenced by their dielectric environment. In this paper the influence of a nondispersive and lossless inhomogeneous dielectric on embedded or nearby resonant atoms is studied. Quantum theory is used both to describe the light and the atoms. As we focus on the effects of the inhomogeneous dielectric, the atoms are modeled simply as quantum harmonic oscillators in their ground states or first excited states, with fixed dipole orientations. To be sure, in choosing this model we neglect optical saturation effects of the atoms. The dielectric is described macroscopically in terms of a real-valued relative dielectric function $\varepsilon(\mathbf{r})$, the form of which will be left arbi-

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trary. The precise measurement of two-atom superradiance in free space as a function of distance is a fundamental test for quantum electrodynamics [16]. The effects calculated here are a test for macroscopic quantization theories for inhomogeneous dielectrics.

Like single-atom emission [19], two-atom superradiance will be modified in the close vicinity of a mirror, or inside an optical cavity [20]. Strong modifications of superradiance are also predicted for photonic crystals, dielectrics with periodic refractive-index variations on the scale of the wavelength of light [21–23]. Large effects are predicted in the so-called isotropic model for a photonic crystal, which is really a toy model in the sense that all local and orientational inhomogeneities of the electromagnetic field are neglected. In a real photonic crystal, two-atom superradiance is expected to sensitively depend on the coordinates of both atoms. The present formalism is valid for an arbitrary real dielectric function and encompasses the interesting special cases just mentioned.

Atoms that exhibit superradiance interact strongly enough to share and exchange the optical excitation before emission. The more common and better studied situation for resonant atoms in a dielectric is that the interaction between the atoms is weak compared to interactions with baths that the individual atoms have. Then the optical excitations are transferred irreversibly from donor to acceptor atoms via a process called "resonance energy transfer," as described by Förster theory [24] and its modern generalizations [25]. Resonance energy transfer is influenced by the dielectric environment. For example, calculations show that two-atom interactions can be strongly influenced by an optical microcavity [26–28], since the cavity modes with eigenfrequencies close or equal to the atomic transition frequency play a dominant role. Indeed, experiments have shown that the interatomic (dipole-dipole) interaction is increased when the atoms are placed in a cavity at positions where resonant optical modes have their maxima [29]. In another interesting experiment, Förster excitation transfer is found to scale linearly with the local optical density of states at the donor position [30]. Although we focus on superradiance, the formalism in the present work is quite general and can also be used as a quantum electrodynamical foundation for the study of energy transfer processes in inhomogeneous media. Recent progress in this direction can be found in [31,32].

Often in quantum optics an "all-matter" picture is employed, where the dynamics of the electromagnetic field is integrated out, for example in the optical Bloch equations [17,33]. Here instead we treat spontaneous emission and superradiance in an "all-light" picture, which is convenient when studying the effect of the dielectric. A multiple-scattering theory is set up in which the atoms show up both as sources and as scatterers of light. It is known that superradiance can be viewed as caused by multiple-scattering interactions [34–36]. Light scattered off a collection of atoms will show multiatom resonances and cooperative effects, also due to multiple scattering. In quantum scattering theory such resonances appear as well and sometimes are called "proximity resonances" [37,38].

The concept of a point scatterer proved very fruitful in the study of multiple-scattering of classical light in free space [39–41]. Here, the point-scattering formalism will be put to use in quantum optics of inhomogeneous dielectrics. Multiple light scattering will be described in terms of Green functions of the medium. The emphasis of the paper will be on formalism, but it ends with an application to superradiance in a model dielectric.

The paper has the following structure: in Sec. II the point-scattering model for interacting guest atoms is introduced. Properties of several types of Green functions of the medium are discussed in Sec. III. Section IV discusses medium-induced modifications of single-atom properties such as spontaneous-emission rates and elastic scattering. The generalization to a finite number of host atoms is discussed in Sec. V. The formalism is applied to two-atom superradiance in Sec. VI, in particular to superradiance near a partially reflecting plane in Sec. VII. Conclusions are drawn in Sec. VIII.

II. ATOMS AS POINT SOURCES AND AS POINT SCATTERERS

A. The Hamiltonian

Consider an inhomogeneous dielectric with relative dielectric function $\varepsilon(\mathbf{r})$ with a finite number N of embedded neutral atoms. The dipole Hamiltonian for this system is the sum of a field part, an atomic part, and an interaction part between field and atoms. More precisely, the Hamiltonian can be found after canonical quantization [11,12] to have the form $\mathcal{H}=\mathcal{H}_F+\mathcal{H}_A+\mathcal{H}_{AF}$, with

$$\mathcal{H}_F = \sum_{\lambda} \hbar\omega_\lambda a_\lambda^\dagger a_\lambda, \tag{1a}$$

$$\mathcal{H}_A = \sum_{m=1}^{N} \hbar\Omega_m b_m^\dagger b_m, \tag{1b}$$

$$\mathcal{H}_{AF} = -\sum_{m=1}^{N} \boldsymbol{\mu}_m \cdot \mathbf{F}(\mathbf{R}_m) = \sum_{m,\lambda} (b_m + b_m^\dagger)(g_{\lambda m} a_\lambda + g_{\lambda m}^* a_\lambda^\dagger). \tag{1c}$$

Notice that there is no direct interaction term between neutral atoms. In a minimal-coupling Hamiltonian, there would have been such a direct coupling term. The situation is analogous to the free-space case [33,42]. The field part $\mathcal{H}_F$ of the Hamiltonian is a sum (or integral) over harmonic oscillators corresponding to the harmonic solutions ("true modes") $\mathbf{f}_\lambda$ of the Maxwell equations for the inhomogeneous dielectric in the absence of the atoms:

$$-\nabla \times \nabla \times \mathbf{f}_\lambda(\mathbf{r}) + \varepsilon(\mathbf{r})(\omega_\lambda/c)^2 \mathbf{f}_\lambda(\mathbf{r}) = 0. \tag{2}$$

For $\omega_\lambda \neq 0$, these modes are generalized transverse, which means that $\nabla\cdot[\varepsilon(\mathbf{r})\mathbf{f}_\lambda(\mathbf{r})]\equiv 0$. Their orthonormality condition reads $\int d\mathbf{r}\varepsilon(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r})\cdot\mathbf{f}_{\lambda'}(\mathbf{r}) = \delta_{\lambda\lambda'}$, where * denotes complex conjugation. The modes are complete, in other words they form a basis for the subspace of generalized transverse functions. For free space $[\varepsilon(\mathbf{r})\equiv 1]$ the $\mathbf{f}_\lambda$ are the well-known transverse plane-wave modes.

In the atomic Hamiltonian $\mathcal{H}_A$, the atomic transition frequencies Ω_m and transition dipole moments $\boldsymbol{\mu}_m$ may be all

different, either because the guest atoms are of different species or because identical atoms feel a different environment. The frequencies Ω_m are assumed real, which means that nonradiative broadening is neglected. The atoms are very simply described as harmonic oscillators with frequencies Ω_m. This is a good approximation within a certain frequency range and as long as saturation effects of the upper atomic state can be neglected. The atomic transition dipole moments $\boldsymbol{\mu}_m$ are assumed to be real-valued and to have fixed orientations. This assumption is better for molecules or quantum dots in a solid surrounding than for atoms in the gas phase. For convenience, the name "atoms" will be used for the guests in the dielectric. The operators $b_m^\dagger(t)$ create atomic excitations by annihilating an atom in the ground state while at the same time creating the atom in the excited state.

The total displacement field $\mathbf{D}(\mathbf{r},t)$ is equal to the displacement field $\varepsilon_0\varepsilon(\mathbf{r})\mathbf{E}(\mathbf{r},t)$ of the inhomogeneous medium plus the sum $\Sigma_m\mathbf{P}_m(\mathbf{r},t)$ of the polarization fields produced by the guest atoms. In the dipole approximation, these polarization fields have the form

$$\mathbf{P}_m(\mathbf{r},t) = \delta(\mathbf{r}-\mathbf{R}_m)\mathbf{P}_m(t) = \delta(\mathbf{r}-\mathbf{R}_m)\boldsymbol{\mu}_m[b_m(t)+b_m^\dagger(t)]. \tag{3}$$

In the dipole interaction term $\mathcal{H}_{AF}$ of the Hamiltonian, a field called $\mathbf{F}$ was introduced that is an abbreviation of

$$\mathbf{F}(\mathbf{r},t) \equiv \mathbf{D}(\mathbf{r},t)/[\varepsilon_0\varepsilon(\mathbf{r})]. \tag{4}$$

Atomic dipoles couple to this field $\mathbf{F}(\mathbf{r},t)$ [11,12]. It is equal to the electric field operator $\mathbf{E}(\mathbf{r},t)$ everywhere, except at the positions $\mathbf{R}_m$ of the guests, since the guest dipoles couple to fields in which their own polarization fields are included. For free space this self-interaction in the dipole coupling is known [42]. The mode expansion of the field $\mathbf{F}(\mathbf{r},t)$ has a simple form, being the sum of a positive-frequency part $\mathbf{F}^{(+)}(\mathbf{r},t)$ containing only annihilation operators and its Hermitian conjugate $\mathbf{F}^{(-)}(\mathbf{r},t)$, where

$$\mathbf{F}^{(+)}(\mathbf{r},t) = i\sum_\lambda \sqrt{\frac{\hbar\omega_\lambda}{2\varepsilon_0}}a_\lambda(t)\mathbf{f}_\lambda(\mathbf{r}). \tag{5}$$

In the absence of the atoms, the time dependence of the annihilation operators in Eq. (5) would be harmonic and $\mathbf{F}(\mathbf{r},t)$ would be equal to the electric field $\mathbf{E}^{(0)}(\mathbf{r},t)$. Here and below, the superscript (0) denotes the absence of guest atoms in the inhomogeneous dielectric. For convenience, coupling constants between atom m and optical mode λ in Eq. (1c) are defined as

$$g_{\lambda m} = -i\sqrt{\frac{\hbar\omega_\lambda}{2\varepsilon_0}}\boldsymbol{\mu}_m\cdot\mathbf{f}_\lambda(\mathbf{R}_m). \tag{6}$$

Notice that the coupling constants $g_{\lambda m}$ are zero for (longitudinal) modes corresponding to $\omega_\lambda=0$. It is by a convenient choice of gauge that the longitudinal modes are decoupled from the atoms in the Hamiltonian (1).

B. Derivation of Lippmann-Schwinger equation

The goal of this section is to derive a Lippmann-Schwinger equation for the field $\mathbf{F}$ inside the inhomogeneous dielectric in the presence of the N guest atoms, by integrating out the atomic dynamics. Heisenberg's equation of motion leads to the following equations of motion for the field operators:

$$\dot{a}_\lambda = -i\omega_\lambda a_\lambda - (i/\hbar)\sum_m g_{\lambda m}^*(b_m+b_m^\dagger), \tag{7a}$$

$$\dot{a}_\lambda^\dagger = i\omega_\lambda a_\lambda^\dagger + (i/\hbar)\sum_m g_{\lambda m}(b_m+b_m^\dagger). \tag{7b}$$

(The dot denotes the time derivative; explicit time dependence of the operators is henceforth dropped.) The field operators are coupled to the atomic operators and the operators of atom m satisfy the equations

$$\dot{b}_m = -i\Omega_m b_m - (i/\hbar)\sum_\lambda (g_{\lambda m}a_\lambda + g_{\lambda m}^* a_{\lambda m}^\dagger), \tag{8a}$$

$$\dot{b}_m^\dagger = i\Omega_m b_m^\dagger + (i/\hbar)\sum_\lambda (g_{\lambda m}a_\lambda + g_{\lambda m}^* a_{\lambda m}^\dagger). \tag{8b}$$

Now take the Laplace transform (or one-sided Fourier transform) of the equations of motion. The transform will have the argument $-i\omega$, for example $b_m(\omega)\equiv\int_0^\infty dt\, e^{i\omega t}b_m(t)$. Here and in the following the frequency ω is assumed to contain an infinitesimally small positive imaginary part so that the transform is well-defined. The equations are algebraic after the transformation.

Also in Fourier language, the equations for the frequency-dependent atomic operators become

$$b_m(\omega) = \frac{ib_m(t=0)}{\omega-\Omega_m} + \frac{\hbar^{-1}}{\omega-\Omega_m}\sum_\lambda [g_{\lambda m}a_\lambda(\omega) + g_{\lambda m}^* a_{\lambda m}^\dagger(\omega)], \tag{9a}$$

$$b_m^\dagger(\omega) = \frac{ib_m^\dagger(t=0)}{\omega+\Omega_m} - \frac{\hbar^{-1}}{\omega+\Omega_m}\sum_\lambda [g_{\lambda m}a_\lambda(\omega) + g_{\lambda m}^* a_{\lambda m}^\dagger(\omega)]. \tag{9b}$$

In obtaining these equations, it was assumed that at time zero, the annihilation operators $a_\lambda(t)$ coincide with the $a_\lambda^{(0)}(t)$, the operators in the absence of the guest atoms. The latter operators have the simple harmonic time dependence $\dot{a}_\lambda^{(0)}(t)+i\omega_\lambda a_\lambda^{(0)}(t)=0$, the transform of which becomes $-i(\omega-\omega_\lambda)a_\lambda^{(0)}(\omega)=a_\lambda^{(0)}(t=0)$ after a partial integration. Notice that $b_m(\omega)$ and $b_m^\dagger(\omega)$ in Eq. (9) are defined as the transforms of $b_m(t)$ and $b_m^\dagger(t)$, respectively. The time-dependent operators are Hermitian conjugates ($b_m^\dagger(t)\equiv[b_m(t)]^\dagger$), but the frequency-dependent operators are not ($b_m^\dagger(\omega)\neq[b_m(\omega)]^\dagger$).

The right-hand sides of Eqs. (9a) and (9b) will now be used to replace $b_m(\omega)$ and $b_m^\dagger(\omega)$ in the Laplace transforms of Eqs. (7a) and (7b) for the field operators. In doing this, the atomic dynamics is integrated out. One obtains for the frequency-dependent annihilation and creation operators of the electromagnetic field

$$a_\lambda(\omega) = a_\lambda^{(0)}(\omega) + \frac{i\hbar^{-1}}{\omega-\omega_\lambda}\sum_m g_{\lambda m}^*\left[\frac{b_m(0)}{\omega-\Omega_m} + \frac{b_m^\dagger(0)}{\omega+\Omega_m}\right], + \frac{\hbar^{-2}}{\omega-\omega_\lambda}\sum_{m,\lambda'}\frac{2g_{\lambda m}^*\Omega_m}{\omega^2-\Omega_m^2}[g_{\lambda' m}a_{\lambda'}(\omega) + g_{\lambda' m}^* a_{\lambda'}^\dagger(\omega)] \tag{10a}$$

$$a_\lambda^\dagger(\omega) = a_\lambda^{(0)\dagger}(\omega) - \frac{i\hbar^{-1}}{\omega+\omega_\lambda}\sum_m g_{\lambda m}\left[\frac{b_m(0)}{\omega-\Omega_m} + \frac{b_m^\dagger(0)}{\omega+\Omega_m}\right] - \frac{\hbar^{-2}}{\omega+\omega_\lambda}\sum_{m,\lambda'}\frac{2g_{\lambda m}\Omega_m}{\omega^2-\Omega_m^2}[g_{\lambda' m}a_{\lambda'}(\omega) + g_{\lambda' m}^* a_{\lambda'}^\dagger(\omega)]. \tag{10b}$$

The optical modes are no longer independent because of the interaction with the atoms. The three terms in the right-hand sides of Eqs. (10a) and (10b) can be related to three reasons why there can be light in mode λ: first, because there is light in the undisturbed mode that has not "seen" the atom; secondly, because the atom can emit light into the mode λ; the third term describes transitions of light in and out of the mode λ to and from modes λ′, due to scattering off one of the guest atoms. Since the relations (10) are implicit rather than explicit solutions for the operators, the identification of terms in the equations with scattering and emission processes can only be approximate.

The results (10) for the creation and annihilation operators can be directly used with Eq. (5) to find the following equation for the field **F**

$$\mathbf{F}(\mathbf{r},\omega) = \mathbf{E}^{(0)}(\mathbf{r},\omega) \tag{11a}$$

$$+\sum_m \mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{S}_m(\omega) \tag{11b}$$

$$+\sum_m \mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{V}_m(\omega)\cdot\mathbf{F}(\mathbf{R}_m,\omega). \tag{11c}$$

This is the central result of this paper. It is an exact Lippmann-Schwinger equation and it describes the resonant scattering off and emission by guest atoms inside an inhomogeneous dielectric, both for strong and for weak atom-field interactions. The equation has an undisturbed term (11a), a source term (11b), and a scattering term (11c).

The elements of Eq. (11) must still be explained. The operator $\mathbf{E}^{(0)}(\mathbf{r},\omega)$ is the electric field in the absence of the atoms, with both the positive and negative frequency parts. The atomic source operators $\mathbf{S}_m(\omega)$ are vectors that have the form $\hat{\boldsymbol{\mu}}_m S_m(\omega)$, where $\hat{\boldsymbol{\mu}}_m$ denotes the unit vector in the direction of the atomic dipole moment $\boldsymbol{\mu}_m$ and

$$S_m(\omega) \equiv \left(\frac{-i\mu_m\omega^2}{\varepsilon_0 c^2}\right)\left[\frac{b_m(0)}{\omega-\Omega_m} + \frac{b_m^\dagger(0)}{\omega+\Omega_m}\right]. \tag{12}$$

Notice that $\mathbf{S}_m$ features the atomic creation and annihilation operators at the initial time zero: in quantum optics, the atomic variables cannot be completely integrated out in an "all-light" picture.

The optical potentials $\mathbf{V}_m(\omega)$ produced by the atoms are dyadics equal to $\hat{\boldsymbol{\mu}}_m V_m(\omega)\hat{\boldsymbol{\mu}}_m$, where

$$V_m(\omega) \equiv \left(\frac{\mu_m^2\omega^2}{\hbar\varepsilon_0 c^2}\right)\left(\frac{2\Omega_m}{\omega^2-\Omega_m^2}\right). \tag{13}$$

Both the sources and the potentials have resonances at frequencies $\pm\Omega_m$. Potentials $V_m(\omega)$ are sometimes rewritten as $-(\omega/c)^2$ times a "bare polarizability" $\alpha_{Bm}(\omega)$ [40]. In the present case, the bare polarizabilities are real (except exactly on resonance) and they change sign when going through their resonances at Ω_m; the resonances are infinitely sharp because all possible nonradiative decay processes are neglected; the polarizability is called "bare" because it does not (and should not) contain radiative broadening of its resonance (but see Sec. IV).

The last undefined factor in Eq. (11) is the dyadic quantity **K** which is given by

$$\mathbf{K}(\mathbf{r},\mathbf{r}',\omega) \equiv c^2\sum_\lambda \frac{\mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')}{(\omega^2-\omega_\lambda^2)}\frac{\omega_\lambda^2}{\omega^2}. \tag{14}$$

Usually, in a Lippmann-Schwinger equation one finds the Green function (called **G**) of a medium where we now find the dyadic **K**. Interestingly, **K** turns out to be different from **G**, even for free space, as will be studied in Sec. III. All the elements of Eq. (11) have now been defined.

Another important field operator for the medium is the vector potential **A**. The magnetic field **B** equals $\boldsymbol{\nabla}\times\mathbf{A}$. In the canonical quantization theories [11,12] upon which our Hamiltonian (1) is based, the generalized Coulomb gauge is chosen, which means that **A** is generalized transverse. Its expansion in terms of the normal modes is given below. With Eq. (10) this leads to

$$\mathbf{A}(\mathbf{r},\omega) \equiv \sum_\lambda \sqrt{\frac{\hbar}{2\varepsilon_0\omega_\lambda}}[a_\lambda(\omega)\mathbf{f}_\lambda(\mathbf{r}) + a_\lambda^\dagger(\omega)\mathbf{f}_\lambda^*(\mathbf{r})] \tag{15a}$$

$$= \mathbf{A}^{(0)}(\mathbf{r},\omega) + \frac{1}{i\omega}\sum_m \mathbf{G}^T(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{S}_m(\omega) + \frac{1}{i\omega}\sum_m \mathbf{G}^T(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{V}_m(\omega)\cdot\mathbf{F}(\mathbf{R}_m,\omega). \tag{15b}$$

Analogously to Eq. (11), an undisturbed term, a source term and a scattering term can be identified for the vector potential.

A difference between Eq. (11) for the field **F** and Eq. (15b) for **A** is that only the former is a Lippmann-Schwinger equation and that **A** immediately follows from the solution of **F**, rather than *vice versa*. In a minimal-coupling formalism, one would find a Lippmann-Schwinger equation for the vector potential instead. Another important difference between the equations for the two fields is that in Eq. (15b) for **A** the generalized transverse Green function $\mathbf{G}^T$ appears, rather than the dyadic **K** of Eq. (11). Definitions of and relations between $\mathbf{G}, \mathbf{G}^T$, and **K** will be studied shortly, in Sec. III.

Often, Lippmann-Schwinger equations are derived in "all-light" formalisms that start with a given optical potential as a perturbation. Here instead, the approach started one level deeper and the optical potential $\mathbf{V}_m$ is output rather than input. An important feature in Eq. (11) is that the atoms are not only point scatterers (potentials), but also point sources for light. Both appear as two sides of the same coin in one equation. Solutions for the equation will be discussed shortly in Sec. IV for one atom and in Sec. V for several atoms.

III. GREEN FUNCTIONS OF THE MEDIUM

The dyadic quantities $\mathbf{K}$ and $\mathbf{G}^T$ will now be related to the Green function of the medium. The (full) Green tensor $\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)$ of an inhomogeneous medium characterized by the dielectric function $\varepsilon(\mathbf{r})$ is the solution of the wave equation

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)+\varepsilon(\mathbf{r})(\omega/c)^2\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)=\delta(\mathbf{r}-\mathbf{r}')\mathbf{I}, \tag{16}$$

where the right-hand side is the ordinary Dirac delta function times the unit tensor.

For a discussion of $\mathbf{G}^T$, it is useful to first introduce the concept of a generalized transverse delta function [5,12]. (For comparison, Green and delta functions of a homogeneous medium are given in the Appendix.) A generalized transverse delta function $\boldsymbol{\delta}_\varepsilon^T$ (a distribution) can be defined in terms of the mode functions $\mathbf{f}_\lambda$ [see Eq. (2)]:

$$\boldsymbol{\delta}_\varepsilon^T(\mathbf{r},\mathbf{r}')\equiv\sum_\lambda \mathbf{f}_\lambda^*(\mathbf{r})\mathbf{f}_\lambda(\mathbf{r}')\varepsilon(\mathbf{r}'). \tag{17}$$

Now $\boldsymbol{\delta}_\varepsilon^T$ has the projection property $\int d\mathbf{r}_1\,\overline{\boldsymbol{\delta}_\varepsilon^T}(\mathbf{r}_1,\mathbf{r})\cdot\mathbf{X}^T(\mathbf{r}_1)=\mathbf{X}^T(\mathbf{r})$ for all (ordinary) transverse vector fields $\mathbf{X}^T$. The bar in $\overline{\boldsymbol{\delta}_\varepsilon^T}$ denotes the transpose. The same projection can be applied to Eq. (16). In doing so, the transverse double-curl term is projected onto itself. The generalized transverse Green function $\mathbf{G}^T$ can now be defined such that $\varepsilon(\mathbf{r})\mathbf{G}^T(\mathbf{r},\mathbf{r}',\omega)$ equals the projection $\int d\mathbf{r}_1\overline{\boldsymbol{\delta}_\varepsilon^T}(\mathbf{r}_1,\mathbf{r})\cdot[\varepsilon(\mathbf{r}_1)\mathbf{G}(\mathbf{r}_1,\mathbf{r}')]$. The projection then leads to the following equation for $\mathbf{G}^T$:

$$-\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)+\varepsilon(\mathbf{r})(\omega/c)^2\mathbf{G}^T(\mathbf{r},\mathbf{r}',\omega)=\overline{\boldsymbol{\delta}_\varepsilon^T}(\mathbf{r}',\mathbf{r}). \tag{18}$$

Notice that $\mathbf{G}$ rather than $\mathbf{G}^T$ appears in the first term. Furthermore, a longitudinal Green function $\mathbf{G}^L$ can be defined as $\mathbf{G}-\mathbf{G}^T$. By taking the difference of Eq. (16) and Eq. (18) one can see that $\mathbf{G}^L$ has the form

$$\mathbf{G}^L(\mathbf{r},\mathbf{r}')\equiv\frac{1}{\varepsilon(\mathbf{r})(\omega/c)^2}[\delta(\mathbf{r}-\mathbf{r}')\mathbf{I}-\overline{\boldsymbol{\delta}_\varepsilon^T}(\mathbf{r}',\mathbf{r})] \tag{19a}$$

$$\equiv\frac{1}{\varepsilon(\mathbf{r})(\omega/c)^2}\overline{\boldsymbol{\delta}_\varepsilon^L}(\mathbf{r}',\mathbf{r}), \tag{19b}$$

In equality (19b) the generalized longitudinal delta function $\boldsymbol{\delta}_\varepsilon^L$ was defined as the difference between the ordinary Dirac and the generalized transverse delta function, so that $\boldsymbol{\delta}_\varepsilon^T+\boldsymbol{\delta}_\varepsilon^L=\delta\mathbf{I}$. We called $\mathbf{G}^L$ the longitudinal Green function, but it is not self-evident that for every inhomogeneous dielectric $\mathbf{G}^L$ is longitudinal indeed. Proofs that $\int d\mathbf{r}'\mathbf{G}^L(\mathbf{r},\mathbf{r}')\cdot\mathbf{X}^T(\mathbf{r}')=0$ and also that $\int d\mathbf{r}\,\mathbf{X}^T(\mathbf{r})\cdot\mathbf{G}^L(\mathbf{r},\mathbf{r}')=0$ can be found with the help of Eqs. (32a) and (32b) of Ref. [12], respectively. Then, since $\mathbf{G}^L$ is longitudinal, $\mathbf{G}$ in Eq. (18) can be replaced by $\mathbf{G}^T$. Hence the projection of Eq. (16) leads to a unique defining equation for $\mathbf{G}^T$.

From Eqs. (2) and (18), it follows that the generalized transverse Green tensor $\mathbf{G}^T$ has the mode expansion

$$\mathbf{G}^T(\mathbf{r},\mathbf{r}',\omega)=c^2\sum_\lambda\frac{\mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')}{(\omega+i\eta)^2-\omega_\lambda^2}. \tag{20}$$

In this manifestly generalized transverse form, $\mathbf{G}^T$ appeared in Eq. (15b) for the vector potential. In the denominator of Eq. (20) we have for once made explicit the positive and infinitesimally small imaginary part of the frequency ω, through the term $i\eta$. With the positive sign of the imaginary part, Eq. (20) is the causal Green function which transformed back to the time-domain gives a Green function $\mathbf{G}^T(\mathbf{r},\mathbf{r}',t-t_0)$ which is nonzero only for positive time differences $(t-t_0)$.

We are now in the position to rewrite and interpret the dyadic $\mathbf{K}$ [see Eq. (14)] that appears in the Lippmann-Schwinger equation (11) for the field $\mathbf{F}$:

$$\mathbf{K}(\mathbf{r},\mathbf{r}',\omega)=c^2\sum_\lambda\frac{\mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')}{\omega^2-\omega_\lambda^2}-(c/\omega)^2\sum_\lambda\mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')$$

$$=\mathbf{G}^T(\mathbf{r},\mathbf{r}',\omega)-\frac{1}{\varepsilon(\mathbf{r})(\omega/c)^2}\overline{\boldsymbol{\delta}_\varepsilon^T}(\mathbf{r}',\mathbf{r}). \tag{21}$$

It consists of the generalized transverse Green function (20) and a term proportional to the transpose of the generalized transverse delta function, $\overline{\boldsymbol{\delta}_\varepsilon^T}$ (17). Both terms are medium-dependent. Note that $\mathbf{K}$ is generalized transverse in its variable $\mathbf{r}$. If only because of this property, $\mathbf{K}$ is not equal to the total Green function (16). Nevertheless, the definition (19b) of the longitudinal Green function can be used to rewrite $\mathbf{K}$ as

$$\mathbf{K}(\mathbf{r},\mathbf{r}',\omega)=\mathbf{G}(\mathbf{r},\mathbf{r}',\omega)-\frac{1}{\varepsilon(\mathbf{r})(\omega/c)^2}\delta(\mathbf{r}-\mathbf{r}')\mathbf{I}. \tag{22}$$

According to this identity, the dyadic $\mathbf{K}$ differs from the full Green function of the medium only when its two position arguments $\mathbf{r}$ and $\mathbf{r}'$ coincide. Although different from $\mathbf{G}$, the quantity $\mathbf{K}$ will also be called a Green function. The occurrence of $\mathbf{K}$ rather than $\mathbf{G}$ in the Lippmann-Schwinger equation will be discussed further in the Appendix, where the volume-integrated electric field around an atom is calculated.

IV. SINGLE-ATOM PROPERTIES ALTERED BY THE MEDIUM

A. Solution of the LS equation

An atom in a group of atoms in an inhomogeneous dielectric will have different properties as compared to free space, because of the dielectric and because of the other atoms. In

this section the effect of the medium on the individual atoms will be considered. The next and major step, in Sec. V, will be to study some effects that the medium-modified atoms can have on each other.

Assume that in the dielectric there is only one guest atom present with dipole moment $\boldsymbol{\mu}$ and transition frequency Ω. The effect of the medium on the scattering and emission properties of the atom can be found by solving Eq. (11) exactly by successive iterations

$$\mathbf{F} = [\mathbf{E}^{(0)} + \mathbf{K}\cdot\mathbf{S}] + \mathbf{K}\cdot\mathbf{V}\cdot[\mathbf{E}^{(0)} + \mathbf{K}\cdot\mathbf{S}] + \mathbf{K}\cdot\mathbf{V}\cdot\mathbf{K}\cdot\mathbf{V}\cdot[\mathbf{E}^{(0)} + \mathbf{K}\cdot\mathbf{S}] + \cdots. \tag{23}$$

In this equation, $\mathbf{K}$ and $\mathbf{V}$ are classical quantities, whereas $\mathbf{F}, \mathbf{E}^{(0)}$, and $\mathbf{S}$ are quantum mechanical operators. The infinite series of multiple-scattering terms can be summed to give

$$\mathbf{F}(\mathbf{r},\omega) = \mathbf{E}^{(0)}(\mathbf{r},\omega) + \mathbf{F}_{\text{scat}}(\mathbf{r},\omega) + \mathbf{F}_{\text{source}}(\mathbf{r},\omega), \tag{24}$$

where, as before, $\mathbf{E}^{(0)}(\mathbf{r},\omega)$ is the electric-field operator of the inhomogeneous medium in the absence of the guest atoms.

The operator $\mathbf{F}_{\text{scat}}(\mathbf{r},\omega)$ in Eq. (24) describes light that is scattered by the guest atom at position $\mathbf{R}$ and it has the form

$$\mathbf{F}_{\text{scat}}(\mathbf{r},\omega) = \mathbf{K}(\mathbf{r},\mathbf{R},\omega)\cdot\mathbf{T}(\omega)\cdot\mathbf{E}^{(0)}(\mathbf{R},\omega), \tag{25}$$

with the single-atom T-matrix defined by

$$\mathbf{T}(\omega) = \hat{\boldsymbol{\mu}}T(\omega)\hat{\boldsymbol{\mu}} = \hat{\boldsymbol{\mu}}\left[\frac{V(\omega)}{1-\hat{\boldsymbol{\mu}}\cdot\mathbf{K}(\mathbf{R},\mathbf{R},\omega)\cdot\hat{\boldsymbol{\mu}}V(\omega)}\right]\hat{\boldsymbol{\mu}}. \tag{26}$$

The T-matrix is sometimes written as $-(\omega/c)^2$ times a dynamical polarizability $\alpha(\omega)$ [compare with Eq. (13)] and both depend on the atomic position inside the inhomogeneous dielectric. The expectation value of the scattered field (25) only depends on the initial quantum state of the light (through the term $\mathbf{E}^{(0)}$). Unlike for a two-level atom, the light-scattering properties of a harmonic-oscillator atom do not depend on the atomic excitation. The scattering process can be read from right to left in the right-hand side of Eq. (25): light $\mathbf{E}^{(0)}$ that has not yet seen the atom scatters off the atom (as described by $\mathbf{T}$), and the scattered part of the light propagates through the dielectric as described by $\mathbf{K}$.

Finally, there is in Eq. (24) the source field

$$\mathbf{F}_{\text{source}}(\mathbf{r},\omega) = \mathbf{K}(\mathbf{r},\mathbf{R},\omega)\cdot\mathbf{S}(\omega) + \mathbf{K}(\mathbf{r},\mathbf{R},\omega)\cdot\mathbf{T}(\omega)\cdot\mathbf{K}(\mathbf{R},\mathbf{R},\omega)\cdot\mathbf{S}(\omega). \tag{27}$$

Expectation values of the source field $\mathbf{F}_{\text{source}}$ only depend on the initial atomic state. Notice that the same T-matrix that shows up in the scattered field (25) also appears in the source field (27). Light emitted by an atomic point source will be studied further in Sec. IV B and scattered light in Sec. IV C.

B. Light emitted by a point source

The source field (27) can be rewritten as

$$\mathbf{F}_{\text{source}}(\mathbf{r},\omega) = \frac{\mathbf{K}(\mathbf{r},\mathbf{R},\omega)\cdot\mathbf{S}(\omega)}{1-\hat{\boldsymbol{\mu}}\cdot\mathbf{K}(\mathbf{R},\mathbf{R},\omega)\cdot\hat{\boldsymbol{\mu}}V(\omega)}, \tag{28}$$

with $\mathbf{S}(\omega)$ as defined in Eq. (12). The time dependence of the source field at the position $\mathbf{r}$ due to the presence of the source at $\mathbf{R}$ follows from the inverse Laplace transform of Eq. (28),

$$\mathbf{F}_{\text{source}}(\mathbf{r},t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega\, e^{-i\omega t}\mathbf{F}_{\text{source}}(\mathbf{r},\omega). \tag{29}$$

This integral cannot be evaluated further without the explicit knowledge of the Green function $\mathbf{K}$. The source field decays in time due to spontaneous emission by the atom. The decay rate can be found by multiplying numerator and denominator in Eq. (28) by $(\omega^2-\Omega^2)$ and by realizing that the zeros $\omega=\Omega_1(\Omega)$ of $\omega^2-\Omega^2-2\Omega X(\omega)$ with

$$X(\omega) \equiv \hat{\boldsymbol{\mu}}\cdot\mathbf{K}(\mathbf{R},\mathbf{R},\omega)\cdot\hat{\boldsymbol{\mu}}[\mu^2\omega^2/(\hbar\varepsilon_0 c^2)] \tag{30}$$

are the frequency poles of the source field. Until now, our solution is exact. At this point we make a pole approximation, which is only valid if the atom-field coupling is weak. The pole approximation gives $\Omega_1=\Omega+X(\Omega)$, with $X(\Omega)$ the difference between the dressed resonance frequency $\Omega_1(\Omega)$ and the bare atomic resonance frequency Ω. The exponential (amplitude) spontaneous-decay rate is

$$\Gamma/2 \equiv -\operatorname{Im} X(\Omega). \tag{31}$$

The decay rate of the intensity of the field is Γ. It is nonnegative by definition of $\mathbf{K}$ in Eq. (14). The delta function term in $\mathbf{K}$ (22) and the longitudinal Green function $\mathbf{G}^L(\mathbf{R},\mathbf{R},\Omega)$ in Eq. (19a) are real quantities, so that Γ is proportional to the imaginary part of only the generalized transverse Green function $\mathbf{G}^T$. The property that $\mathbf{G}^L$ does not contribute to the spontaneous-emission rate is a generalization of the well-known result for homogeneous dielectrics [43] and it only holds for nonabsorbing dielectrics. Using the mode composition Eq. (20) of $\mathbf{G}^T$, we find $\Gamma=\pi/(\hbar\varepsilon_0)\Sigma_\lambda|\boldsymbol{\mu}\cdot\mathbf{f}_\lambda(\mathbf{r})|^2\omega_\lambda\delta(\Omega-\omega_\lambda)$, the same expression that one also finds from Fermi's golden rule [5]. The decay rate depends both on the atom's position and on its orientation inside the inhomogeneous dielectric. For free space, the imaginary part of $\mathbf{G}_0^T(\mathbf{R},\mathbf{R},\Omega)$ is equal to $-\Omega/(6\pi c)\mathbf{I}$ [see Eq. (A2a)]. This gives the familiar free-space spontaneous-decay rate $\Gamma_0=\mu^2\Omega^3/(3\pi\hbar\varepsilon_0 c^3)$.

The dressed resonance frequency Ω_1 can be written as $\Omega+\Delta'(\Omega)-i\Gamma(\Omega)/2$. Apart from a decay rate there is a frequency shift Δ' that is equal to $\operatorname{Re} X(\Omega)$. For two reasons, Δ' is infinitely large even for free space. First, the delta function term $\delta(\mathbf{r}-\mathbf{R})\mathbf{I}/[\varepsilon(\mathbf{R})(\omega/c)^2]$ in Eq. (22) diverges when $\mathbf{r}$ and $\mathbf{R}$ are equal. This self-interaction term is medium-dependent through the factor $\varepsilon(\mathbf{R})$, but here and in the following we assume that guest atoms are electronically well separated from the dielectric medium, so that an empty-cavity model applies where the relative dielectric function is equal to unity at the position of the guest atom [12]. The second reason why Δ' diverges is well known for free space: $\mathbf{G}_0(\mathbf{r},\mathbf{R},\omega)$ diverges when $\mathbf{r}$ approaches $\mathbf{R}$ [see Eqs. (A2a) and (A2b)].

By a procedure called mass renormalization, the combined radiative shift in free space becomes finite, see for example [44]. From now on we can assume that Ω is the observable atomic frequency in free space; inside a dielectric, the atomic frequency shifts by an amount Δ that is given by the real part of $[X(\omega)-X_0(\Omega)]$, or in terms of the Green functions

$$\Delta = \hat{\boldsymbol{\mu}} \cdot \mathrm{Re}[\mathbf{G}(\mathbf{R},\mathbf{R},\Omega) - \mathbf{G}_0(\mathbf{R},\mathbf{R},\Omega)] \cdot \hat{\boldsymbol{\mu}} \left(\frac{\mu^2 \Omega^2}{\hbar \varepsilon_0 c^2} \right). \tag{32}$$

The shift depends on the atomic position and dipole orientation. Notice that the full Green function is needed to determine the line shift, whereas for the decay rate it sufficed to know $\mathbf{G}^T$.

The position-dependent radiative shifts are a mechanism of inhomogeneous broadening of the detected light. Electronic shifts usually dominate inhomogeneous broadening. Experimentally it will be hard to single out radiative shifts (a photonic effect) from electronic line shifts (due to changes in the atomic wave functions inside the medium).

C. Light scattered by a point scatterer

In the scattered field of Eq. (25), the atom appears as a point scatterer with an internal resonance in the optical potential $V(\omega)$ and a corresponding resonance in the T-matrix in Eq. (26). The scattered field has frequency poles in the T-matrix (just like the source field), but it also has poles for every optical mode frequency $\pm\omega_\lambda$ (unlike the source field). The time-dependence of the scattered field can be understood by separating the frequency poles (straightforward, but not spelled out here), again followed by an inverse Laplace transformation. In the following, place the atom in the origin. For the part of $\mathbf{F}_{\mathrm{scat}}$ featuring the annihilation operators, one finds

$$\begin{aligned}\mathbf{F}^{(+)}_{\mathrm{scat}}(\mathbf{r},t) = \sum_\lambda & \frac{-\mu^2}{2\pi\hbar\varepsilon_0 c^2} \sqrt{\frac{\hbar\omega_\lambda}{2\varepsilon_0}} a^{(0)}_\lambda(0) \mathbf{f}_\lambda(\mathbf{0}) \cdot \hat{\boldsymbol{\mu}} \\ & \times \int_{-\infty}^{\infty} d\omega\, \omega^2 e^{-i\omega t} \mathbf{K}(\mathbf{r},\mathbf{0},\omega) \cdot \hat{\boldsymbol{\mu}} \\ & \times \left\{ -\frac{2\Omega}{(\Omega+\Delta)^2 - \omega_\lambda^2 + \Gamma^2/4 - i\omega_\lambda\Gamma} \frac{1}{\omega - \omega_\lambda} \right. \\ & + \frac{\Omega/(\Omega+\Delta)}{\Omega + \Delta - \omega_\lambda - i\Gamma/2} \frac{1}{\omega - \Omega - \Delta + i\Gamma/2} \\ & \left. + \frac{\Omega/(\Omega+\Delta)}{\Omega + \Delta + \omega_\lambda + i\Gamma/2} \frac{1}{\omega + \Omega + \Delta + i\Gamma/2} \right\}. \end{aligned} \tag{33}$$

The negative-frequency part $\mathbf{F}^{(-)}$ of the field equals $[\mathbf{F}^{(+)}]^\dagger$. The three terms between curly brackets in Eq. (33) correspond to different optical processes. The first term describes elastic light scattering by the guest atom inside the inhomogeneous dielectric; the second term has an exponentially decaying time dependence and corresponds to resonance fluorescence; finally, the third term is an exponentially decaying nonresonant term, corresponding to an utterly improbable process that one could call antiresonance fluorescence. In a rotating-wave approximation this process would disappear. After neglecting this third term, all the ω-poles in the integral (33) have positive real parts, so that Eq. (33) can be called the positive-frequency part of the field $\mathbf{F}_{\mathrm{scat}}$.

Now consider the second term in Eq. (33) in more detail. In the resonance fluorescence process the guest atom is excited by light of frequency ω_p, after which the atomic source decays exponentially due to light emission at frequency Ω_s. (In contrast, elastically scattered light oscillates with the pump frequency ω_p.) The fluorescent light has the same position-dependent emission rates $\Gamma(\mathbf{R},\Omega)$ (31) and line shifts $\Delta(\mathbf{R},\Omega)$ (32) as found for spontaneous emission before. A difference between the source-field of Eq. (28) and the fluorescent light in Eq. (33) is that the latter also contains the information how well the pump light that comes in via mode $\lambda=p$ can excite the atom, in the factor $\mathbf{f}_p(0)\cdot\hat{\boldsymbol{\mu}}$. This difference is especially important for inhomogeneous dielectrics, where atoms will be excited easier here than there. And indeed, it is through the process of resonance fluorescence that lifetimes of atoms in dielectric media are usually measured.

In a resonance fluorescence experiment, a light pulse or wave packet passes the atom during a time T. In expression (33), the intensity of resonantly emitted light depends on the expectation value with respect to the quantum state of light at time $t=0$. This can only be a valid description of the process if $T \ll \Gamma^{-1}$, in other words, if the wave packet is so short that it "prepares percussionally the excited state" ([45], p. 97) of the atom at time $t=0$. This is typically the case, even if the medium broadens the excitation pulse: excitation pulses last picoseconds and lifetimes lie in the nanosecond regime.

V. SEVERAL ATOMS AS POINT SOURCES AND SCATTERERS

A. Solution of the LS equation

In Sec. IV it was found how scattering by and emission rates of single atoms are influenced by their dielectric surroundings. In the present section it is studied how the medium-modified atoms can influence each other. The atomic wave functions are assumed not to overlap each other and to be unaffected by the dielectric. The atomic positions can be arbitrary, so we can decide to choose the atoms on a line [46] or on a lattice [47] or at random positions. The general method to solve the Lippmann-Schwinger equation (11) in this more complicated situation is outlined here. In Sec. VI, the formalism will be used to study two-atom superradiance inside an inhomogeneous dielectric medium.

For one atom, the Lippmann-Schwinger equation (11) was solved by summing a series to all orders of the atomic potential. In the present many-atom case all atomic transition dipole moments, orientations, and frequencies are allowed to be different so that also all optical potentials $\mathbf{V}_j$ are different. The LS equation will now be solved by efficiently summing a somewhat more complicated series. Use the abbreviations $\mathbf{F}=\mathbf{F}(\mathbf{r},\omega)$, $\mathbf{F}_m=\mathbf{F}(\mathbf{R}_m,\omega)$, $\mathbf{K}_m=\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)$, $\mathbf{K}_{mn}$

$=\mathbf{K}(\mathbf{R}_m,\mathbf{R}_n,\omega)$, and introduce $\mathbf{F}^{(1)}\equiv\mathbf{E}^{(0)}(\mathbf{r},\omega)+\Sigma_m\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{S}_m(\omega)$. Also, $\mathbf{F}_n^{(1)}$ is shorthand for $\mathbf{F}^{(1)}(\mathbf{R}_n)$. By iteration it follows that the field $(\mathbf{F}-\mathbf{F}^{(1)})$ of Eq. (11) becomes

$$\sum_{n=1}^{N}\mathbf{K}_n\cdot\mathbf{V}_n\cdot\mathbf{F}_n^{(1)}+\sum_{m,n=1}^{N}\mathbf{K}_m\cdot\mathbf{V}_m\cdot\mathbf{K}_{mn}\cdot\mathbf{V}_n\cdot\mathbf{F}_n^{(1)}+\sum_{m,p,n=1}^{N}\mathbf{K}_m\cdot\mathbf{V}_m\cdot\mathbf{K}_{mp}\cdot\mathbf{V}_p\cdot\mathbf{K}_{pn}\cdot\mathbf{V}_n\cdot\mathbf{F}_n^{(1)}+\cdots. \tag{34a}$$

This can conveniently be rewritten in terms of the single-atom T-matrices of Eq. (26) as

$$\sum_{n=1}^{N}\mathbf{K}_n\cdot\mathbf{T}_n\cdot\mathbf{F}_n^{(1)}+\sum_{m,n=1}^{N}\mathbf{K}_m\cdot\mathbf{T}_m\cdot\mathbf{K}'_{mn}\cdot\mathbf{T}_n\cdot\mathbf{F}_n^{(1)}+\sum_{m,p,n=1}^{N}\mathbf{K}_m\cdot\mathbf{T}_m\cdot\mathbf{K}'_{mp}\cdot\mathbf{T}_p\cdot\mathbf{K}'_{pn}\cdot\mathbf{T}_n\cdot\mathbf{F}_n^{(1)}+\cdots. \tag{34b}$$

Here the tensor $\mathbf{K}'_{mn}$ is defined as $(1-\delta_{mn})\mathbf{K}_{mn}$, which by virtue of Eq. (22) is equal to $\mathbf{G}'_{mn}\equiv(1-\delta_{mn})\mathbf{G}_{mn}$. A single-atom T-matrix already sums up all multiple potential-scattering off a single atom, which explains that neighboring T-matrices in terms of this series belong to different atoms. The equivalence of Eqs. (34a) and (34b) can be seen by expanding single-atom T-matrices in terms of single-atom potentials. Now every higher-order term in Eq. (34b) can be constructed from the previous-order term by inserting into the latter the $N\times N$ matrix with (i,j)-elements $\hat{\boldsymbol{\mu}}_i\cdot\mathbf{G}'_{ij}\cdot\mathbf{T}_j\cdot\hat{\boldsymbol{\mu}}_j$. By summing the geometric series of matrices and dropping the abbreviations, it follows that

$$\mathbf{F}(\mathbf{r},\omega)=\mathbf{F}^{(1)}(\mathbf{r},\omega)+\sum_{m,n=1}^{N}\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{T}_{mn}^{(N)}(\omega)\cdot\mathbf{F}^{(1)}(\mathbf{R}_n,\omega), \tag{35}$$

with the N-atom T-matrix

$$\mathbf{T}_{mn}^{(N)}(\omega)=\hat{\boldsymbol{\mu}}_m T_{mn}^{(N)}(\omega)\hat{\boldsymbol{\mu}}_n=\hat{\boldsymbol{\mu}}_m T_m(\omega)M_{mn}^{-1}(\omega)\hat{\boldsymbol{\mu}}_n. \tag{36}$$

The $N\times N$ matrix $\mathbf{M}(\omega)$ is defined as

$$M_{ij}(\omega)=\delta_{ij}-(1-\delta_{ij})\hat{\boldsymbol{\mu}}_i\cdot\mathbf{G}(\mathbf{R}_i,\mathbf{R}_j,\omega)\cdot\hat{\boldsymbol{\mu}}_j T_j(\omega). \tag{37}$$

Equations (36) and (37) neatly sum up infinitely many scattering events which are not described by $\mathbf{G}$. Light propagation in between the scattering off one atom and the next one is described by $\mathbf{G}$ and need not be rectilinear, since $\mathbf{G}$ is the Green function of the inhomogeneous medium.

As before, the total field $\mathbf{F}$ consists of the part $\mathbf{E}^{(0)}$ that has not seen the atoms, a scattered part and a source-field part. The field operator that describes the scattering of light by the N-atom system has the form

$$\mathbf{F}_{\text{scat}}(\mathbf{r},\omega)=\sum_{m,n=1}^{N}\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{T}_{mn}^{(N)}(\omega)\cdot\mathbf{E}^{(0)}(\mathbf{R}_n,\omega). \tag{38}$$

This is a generalization of the single-atom result of Eq. (25). It describes elastic scattering as well as resonance fluorescence off N atoms in an inhomogeneous medium. The expectation value of $\mathbf{F}_{\text{scat}}$ depends on the initial quantum state of light only. Similarly, for the N-atom source-field that only depends on the initial atomic state, we find

$$\mathbf{F}_{\text{source}}(\mathbf{r},\omega)=\sum_{m=1}^{N}\mathbf{K}^{(N)}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{S}_m(\omega), \tag{39}$$

which generalizes Eq. (27). Here, $\mathbf{K}^{(N)}$ is a Green function of the inhomogeneous dielectric including the N atoms:

$$\mathbf{K}^{(N)}(\mathbf{r},\mathbf{r}',\omega)=\mathbf{K}(\mathbf{r},\mathbf{r}',\omega)+\sum_{m,n=1}^{N}\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{T}_{mn}^{(N)}(\omega)\cdot\mathbf{K}(\mathbf{R}_n,\mathbf{r}',\omega). \tag{40}$$

For $\mathbf{r}$ and $\mathbf{r}'$ different from one of the positions $\mathbf{R}_m$, $\mathbf{K}^{(N)}(\mathbf{r},\mathbf{r}',\omega)$ is equal to what one would call the total Green function $\mathbf{G}^{(N)}$ of both the dielectric and its guests. The solution (39) shows that the source-field $\mathbf{K}^{(N)}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{S}_m(\omega)$ that emanates from atom m is influenced by the positions, orientations, dipole moments and resonance frequencies of the $(N-1)$ other atoms. Notice that the same N-atom T-matrix describes the N-atom source fields and scattered fields. The two-atom source field will be studied in Sec. VI.

B. Interatomic interactions

In the results of Sec. V A, interatomic interactions can be identified. Before doing that, we briefly mention possible interatomic interactions that we already neglected or that simply do not occur in our theory. In a minimal-coupling formalism there would be a direct atom-atom interaction in the Hamiltonian. In a multipole formalism, the only direct interaction between neutral atoms is an interatomic polarization energy [42]. Classically, this interaction is zero unless the smallest spheres containing the atomic charges have nonzero overlap [42]. Quantum mechanically, this "contact energy" is negligible unless the interatomic distance is of the order of the size of the atoms such that wave functions overlap. We assumed that the atoms were further apart. Together with the fact that atoms are much smaller than the wavelength of light, this allows us to make the dipole approximation in which atoms are considered as point dipoles. Therefore, direct interactions between the atoms are absent in the dipole Hamiltonian Eq. (1). Our approximations make that the only interatomic interactions that we can find are retarded dipole-dipole interactions, mediated by the electromagnetic field.

Indeed, in Sec. V A interatomic interactions showed up in the N-atom T-matrix as terms proportional to the causal Green tensor of the medium. For two noncoinciding posi-

tions $\mathbf{R}_1$ and $\mathbf{R}_2$, the interaction (with dimension: [frequency]) has the form

$$J_{12} = J(\mathbf{R}_1,\mathbf{R}_2,\omega) = \frac{\mu_1\mu_2\omega^2}{\hbar\varepsilon_0 c^2}\hat{\boldsymbol{\mu}}_1\cdot\mathbf{G}(\mathbf{R}_1,\mathbf{R}_2,\omega)\cdot\hat{\boldsymbol{\mu}}_2$$

$$= \frac{\mu_1\mu_2}{\hbar\varepsilon_0}\sum_\lambda \hat{\boldsymbol{\mu}}_1\cdot\mathbf{f}_\lambda(\mathbf{R}_1)\mathbf{f}_\lambda^*(\mathbf{R}_2)\cdot\hat{\boldsymbol{\mu}}_2\frac{\omega_\lambda^2}{\omega^2-\omega_\lambda^2}. \qquad (41)$$

For the latter identity, Eqs. (14) and (22) were used. Only after making a pole approximation in Sec. VI will it become fully clear why we identify precisely this expression as the dipole-dipole interaction. Modes with eigenfrequencies $\omega_\lambda \equiv 0$ were absent in the dipole interaction (1c) and consequently are absent in the dipole-dipole interaction (41).

The Green function $\mathbf{G}$ can be written as the sum of the generalized transverse Green function $\mathbf{G}^T$ and a longitudinal Green function $\mathbf{G}^L$ [recall Eqs. (19)–(22)]. The dipole-dipole interaction can be split into two analogous parts. The generalized transverse part is

$$J_{\mathrm{gtrans}}(\mathbf{R}_1,\mathbf{R}_2,\omega) = \frac{\mu_1\mu_2\omega^2}{\hbar\varepsilon_0}\sum_\lambda \frac{\hat{\boldsymbol{\mu}}_1\cdot\mathbf{f}_\lambda(\mathbf{R}_1)\mathbf{f}_\lambda^*(\mathbf{R}_2)\cdot\hat{\boldsymbol{\mu}}_2}{\omega^2-\omega_\lambda^2}. \qquad (42a)$$

It is also called the "resonant dipole-dipole interaction" (or RDDI). The strongest contribution to this interaction comes from the modes λ with eigenfrequencies ω_λ near ω, which explains the adjective "resonant." Notice that J_{gtrans} is zero when ω is zero. The other part is the longitudinal dipole-dipole interaction J_{long} that has the mode expansion

$$J_{\mathrm{long}}(\mathbf{R}_1,\mathbf{R}_2) = -\frac{\mu_1\mu_2}{\hbar\varepsilon_0}\sum_\lambda \hat{\boldsymbol{\mu}}_1\cdot\mathbf{f}_\lambda(\mathbf{R}_1)\mathbf{f}_\lambda^*(\mathbf{R}_2)\cdot\hat{\boldsymbol{\mu}}_2. \qquad (42b)$$

Notice that J_{long} is independent of the frequency ω. It is the generalization of the static dipole-dipole interaction that is well known for free space. Both the generalized transverse and the longitudinal dipole-dipole interactions are given here in terms of generalized transverse modes. Both J_{gtrans} and J_{long} are influenced by the medium.

Both $\mathbf{G}^T$ and $\mathbf{G}^L$ have nonretarded dipole terms, so that a change in a source term changes instantaneously the longitudinal and generalized transverse fields elsewhere. It is only their sum that is fully retarded. This is well known for free-space Green tensors (see [48]) and it holds likewise for the Green functions $\mathbf{G}^T_{\mathrm{hom}}$ and $\mathbf{G}^L_{\mathrm{hom}}$ of homogeneous dielectrics as given in the Appendix.

It might seem strange that the longitudinal interaction Eq. (42b) is not given in terms of longitudinal modes. The physical reason is that longitudinal modes do not couple to the atoms in our formalism (see Sec. II A). Still, apart from the generalized transverse solutions $\mathbf{f}_\lambda$ (with $\omega_\lambda \neq 0$) of the wave equation (2), there are also longitudinal solutions $\mathbf{q}_\nu$ (with $\omega_\nu = 0$). There is a mathematical identity that allows one to rewrite the longitudinal interaction Eq. (42b) in terms of longitudinal modes. The identity originates from the fact that the modes $\{\mathbf{f}_\lambda,\mathbf{q}_\nu\}$ together span the entire space of functions $\mathbf{h}$ with $\int d\mathbf{r}\varepsilon(\mathbf{r})|\mathbf{h}(\mathbf{r})|^2 < \infty$. This space consists of a subspace of generalized transverse functions and a longitudinal subspace. The completeness relation in the entire space reads $\Sigma_\lambda \mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')\varepsilon(\mathbf{r}') + \Sigma_\nu \mathbf{q}_\nu(\mathbf{r})\mathbf{q}_\nu^*(\mathbf{r}')\varepsilon(\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}')\mathbf{l}$, with $\mathbf{l}$ the unit tensor. It follows that for $\mathbf{r}\neq\mathbf{r}'$, one can replace $\Sigma_\lambda \mathbf{f}_\lambda(\mathbf{r})\mathbf{f}_\lambda^*(\mathbf{r}')$ in Eq. (42b) by minus the sum $\Sigma_\nu \mathbf{q}_\nu(\mathbf{r})\mathbf{q}_\nu^*(\mathbf{r}')$. Incidentally, the longitudinal modes $\mathbf{q}_\nu$ of the medium are different from the free-space longitudinal modes, because of their different orthogonality relations $\int d\mathbf{r}\varepsilon(\mathbf{r})\mathbf{q}_\nu(\mathbf{r})\cdot\mathbf{q}_{\nu'}^*(\mathbf{r}) = \delta_{\nu\nu'}$.

VI. TWO-ATOM SUPERRADIANCE IN INHOMOGENEOUS MEDIUM

The general results of Sec. V will now be applied to two identical atoms positioned in an inhomogeneous dielectric. Assume that the two atoms have identical electronic transition frequencies Ω and dipole moments $\mu = |\boldsymbol{\mu}|$; their dipole orientations $\hat{\boldsymbol{\mu}}_1$ and $\hat{\boldsymbol{\mu}}_2$ need not be identical. The source field of this two-atom system is [see Eq. (39)]

$$\mathbf{F}_{\mathrm{source}}(\mathbf{r},\omega) = \mathbf{K}^{(2)}(\mathbf{r},\mathbf{R}_1,\omega)\cdot\mathbf{S}_1(\omega) + \mathbf{K}^{(2)}(\mathbf{r},\mathbf{R}_2,\omega)\cdot\mathbf{S}_2(\omega). \qquad (43)$$

The goal is now to calculate the Green function $\mathbf{K}^{(2)}$ of the dielectric including the guest atoms, in terms of the properties of the medium and of the individual atoms.

According to Eq. (40), the Green function $\mathbf{K}^{(2)}$ is known once the T-matrix $T^{(2)}$ (36) is determined; $T^{(2)}$ can be found by inverting the 2×2 matrix M (37), in which the single-atom T-matrices occur that are given in Eq. (26) and the Green function $\mathbf{K}$ of the dielectric in Eq. (22). It follows that the two-atom T-matrix is

$$\mathbf{T}^{(2)} = \frac{1/\beta}{1-T_1J_{12}^2T_2/\beta^2}\begin{pmatrix} \hat{\boldsymbol{\mu}}_1\hat{\boldsymbol{\mu}}_1\beta T_1 & \hat{\boldsymbol{\mu}}_1\hat{\boldsymbol{\mu}}_2 T_1J_{12}T_2 \\ \hat{\boldsymbol{\mu}}_2\hat{\boldsymbol{\mu}}_1 T_2J_{12}T_1 & \hat{\boldsymbol{\mu}}_2\hat{\boldsymbol{\mu}}_2\beta T_2 \end{pmatrix}, \qquad (44)$$

with the dipole-dipole interaction J_{12} defined in Eq. (41) and β as $\mu^2\omega^2/(\hbar\varepsilon_0 c^2)$. Each of the four matrix elements of $\mathbf{T}^{(2)}$ is a dyadic of the same type as the single-atom T-matrix (26). Now abbreviate $\mathbf{K}(\mathbf{r},\mathbf{R}_1,\omega)$ as $\mathbf{K}(\mathbf{r}1)$ and similarly for other terms. The Green function $\mathbf{K}^{(2)}(\mathbf{r}1)$ can be written with Eq. (40) as

$$\mathbf{K}^{(2)}(\mathbf{r}1) = \mathbf{K}(\mathbf{r}1)\cdot[\mathbf{I} + \mathbf{T}_{11}^{(2)}\cdot\mathbf{K}(11) + \mathbf{T}_{12}^{(2)}\cdot\mathbf{K}(21)] + \mathbf{K}(\mathbf{r}2)\cdot[\mathbf{T}_{21}^{(2)}\cdot\mathbf{K}(11) + \mathbf{T}_{22}^{(2)}\cdot\mathbf{K}(21)]. \qquad (45)$$

Use Eq. (44) to rewrite the T-matrix elements of $\mathbf{T}^{(2)}$ in terms of the single-atom T-matrices. The first one of the two parts of the source field (43) is associated with light initially residing in atom 1. This part can be written in terms of single-atom properties as

$$\mathbf{K}^{(2)}(\mathbf{r}1)\cdot\mathbf{S}_1 = \left(\frac{1+T_1X_1/\beta}{1-T_1J_{12}^2T_2/\beta^2}\right)\times[\mathbf{K}(\mathbf{r}1)\cdot\hat{\boldsymbol{\mu}}_1 + \mathbf{K}(\mathbf{r}2)\cdot\hat{\boldsymbol{\mu}}_2T_2J_{21}/\beta]S_1, \qquad (46)$$

with $X_i = X_i(\omega)$ as defined in Eq. (30). The source field has

now been expressed in terms of the T-matrices of the individual atoms, but it is rewarding to break up the T-matrices in parts that depend on the medium alone and parts that depend on the atoms:

$$\mathbf{K}^{(2)}(\mathbf{r}1)\cdot\mathbf{S}_1=\frac{(\omega^2-\Omega^2)S_1[\mathbf{K}(\mathbf{r}1)\cdot\hat{\boldsymbol{\mu}}_1(\omega^2-\Omega^2-2\Omega X_2)+\mathbf{K}(\mathbf{r}2)\cdot\hat{\boldsymbol{\mu}}_2 2\Omega J_{12}]}{(\omega^2-\Omega^2)^2-2\Omega(X_1+X_2)(\omega^2-\Omega^2)+4\Omega^2(X_1X_2-J_{12}^2)}. \tag{47}$$

The denominator carries the important information about the resonance frequencies $\Omega_\pm(\Omega)$ of the two-atom system. There are two resonance frequencies near Ω and two near $-\Omega$. If these resonance frequencies change little due to the electromagnetic coupling with the dielectric, we can replace the frequency dependent functions $X_{1,2}(\omega)$ and $J_{12}(\omega)$ in the above expression by their values in $\omega=\pm\Omega$. This is the pole approximation that we also made for the single atom. We find the two resonance frequencies

$$\Omega_\pm(\Omega)=\Omega+\frac{X_1+X_2}{2}\pm\sqrt{\left(\frac{X_1-X_2}{2}\right)^2+J_{12}^2}. \tag{48}$$

The other two resonance frequencies occur at $-\Omega_\pm^*(\Omega)$, so that all four have negative imaginary parts.

When the atoms are far apart, then J_{12} tends to zero and the two resonance frequencies $\Omega_\pm$ (48) are simply the two single-atom frequencies Ω_1 and Ω_2 with their medium-dependent radiative shifts $\Delta_{1,2}$ and decay rates $\Gamma_{1,2}$. In the other extreme situation, for atoms with parallel dipoles atoms (almost) on top of each other, $-\mathrm{Im}\,\Omega_+$ approaches twice the single-atom amplitude decay rate, whereas $-\mathrm{Im}\,\Omega_-$ has the limiting value zero. Analogous to free space, Ω_+ corresponds to the superradiant state of the two-atom system in the medium, whereas Ω_- is the frequency belonging to the subradiant state.

Now rewrite Eq. (47) as a sum over individual first-order frequency poles. With Eq. (12) one has

$$\begin{aligned}\mathbf{K}^{(2)}(\mathbf{r}1)\cdot\mathbf{S}_1=\sum_\pm&\left(\frac{-i\mu\omega^2}{4\varepsilon_0c^2\Omega_\pm}\right)\\&\times[\mathbf{K}(\mathbf{r}1)\cdot\hat{\boldsymbol{\mu}}_1(1\pm\sin\alpha)\pm\mathbf{K}(\mathbf{r}2)\cdot\hat{\boldsymbol{\mu}}_2\cos\alpha]\\&\times[(\omega+\Omega)b_1(0)+(\omega-\Omega)b_1^\dagger(0)]\\&\times\left(\frac{1}{\omega-\Omega_\pm}-\frac{1}{\omega+\Omega_\pm}\right).\end{aligned} \tag{49}$$

A (complex) angle $\alpha=\alpha(\Omega)$ has been introduced which measures the inhomogeneity of the medium as felt by the two-atom system, in comparison with the atom-atom interaction:

$$\sin\alpha\equiv(\Omega_1-\Omega_2)/\Lambda,\quad\cos\alpha\equiv 2J_{12}/\Lambda, \tag{50}$$

with Λ equal to $\sqrt{(\Omega_1-\Omega_2)^2+4J_{12}^2}$. When the angle α is zero (such as in free space), the atoms are said to be placed at equivalent positions in the medium.

In the expression (48) for the resonance frequencies and in the angle α (50), a driving term and a detuning can be discerned. The driving term is the dipole-dipole interaction J_{12} and it signifies how important the one atom is as a light source for the other. The term $(\Omega_1-\Omega_2)/2$ is a detuning: larger medium-induced local differences felt by the identical atoms make the resonant transfer of a photon between them less probable. The driving term and the detuning have the same physical origin and cannot be changed independently. By bringing the atoms much closer in each other's near field, they will be tuned better and interact stronger at the same time. The outcome of the competition between medium-induced driving and detuning will be studied in an example in Sec. VII.

The time dependence of the source field can now be calculated with an inverse Laplace transformation. Notice that the positive-frequency poles in Eq. (49) have negligible residues in terms proportional to $b_1^\dagger$; similarly, negative-frequency poles hardly contribute to terms involving the annihilation operator $b_1(0)$ and can be neglected as well. The total source field $\mathbf{F}_{\mathrm{source}}(\mathbf{r},t)$ is the field (49) that originates from the initial excitation of the atom labeled 1, accompanied by the source field that originally came from the second atom:

$$\mathbf{F}_{\mathrm{source}}(\mathbf{r},t)=\mathbf{L}_1(\mathbf{r},t)b_1(0)+\mathbf{L}_2(\mathbf{r},t)b_2(0)+\mathrm{H.c.} \tag{51}$$

The vector $\mathbf{L}_1$ can be written as the sum of $\mathbf{L}_{1+}$ and $\mathbf{L}_{1-}$, with

$$\begin{aligned}\mathbf{L}_{1\pm}(\mathbf{r},t)\equiv&\frac{-i\mu}{4\pi\varepsilon_0c^2}\int_{-\infty}^{\infty}d\omega\frac{\omega^2e^{-i\omega t}}{\omega-\Omega_\pm}\frac{\omega+\Omega}{2\Omega_\pm}\\&\times[\mathbf{K}(\mathbf{r},\mathbf{R}_1,\omega)\cdot\hat{\boldsymbol{\mu}}_1(1\pm\sin\alpha)\\&\pm\mathbf{K}(\mathbf{r},\mathbf{R}_2,\omega)\cdot\hat{\boldsymbol{\mu}}_2\cos\alpha].\end{aligned} \tag{52}$$

This is the central result of this section. Vectors $\mathbf{L}_{2\pm}$ can be found by interchanging the indices 1 and 2 in the right-hand side of Eq. (52), which also causes a sign change in $\sin\alpha$ (50).

Equation (52) describes the full time dependence of the source that is excited at time $t=0$. Initially light has been emitted but has not arrived at the detector yet. This initial phase lasts a certain delay time t_d, depending on the optical path length between source and detector. The initial phase is followed by a transient regime, in which light that has chosen the shortest path already arrives at the detector at $\mathbf{r}$, while light that takes a longer path has not arrived yet. The transient regime can be neglected if it lasts much shorter than

the typical atomic decay time, which is usually the case. Assuming the same delay time for both resonance frequencies and for both atoms, we find

$$\mathbf{L}_{1\pm}(\mathbf{r},t) \equiv \frac{-\mu\Omega_{\pm}(\Omega+\Omega_{\pm})}{4\varepsilon_0 c^2}\theta(t-t_d)e^{-i\Omega_{\pm}t} \times [\mathbf{K}(\mathbf{r},\mathbf{R}_1,\Omega)\cdot\hat{\boldsymbol{\mu}}_1(1\pm\sin\alpha) \pm \mathbf{K}(\mathbf{r},\mathbf{R}_2,\Omega)\cdot\hat{\boldsymbol{\mu}}_2\cos\alpha]. \quad (53)$$

In this equation we see that the source amplitudes of the atoms are influenced by their environment. There are overall factors which are equal for both atoms. The atoms differ in that the source amplitude of the light that is finally emitted by the first atom has a factor $(1\pm\sin\alpha)$ while the corresponding factor for the second atom is $\cos\alpha$. The results for the source field can be inserted into the intensity operator [33]

$$I(\mathbf{r},t) = 2\varepsilon_0 c\mathbf{E}^{(-)}(\mathbf{r},t)\cdot\mathbf{E}^{(+)}(\mathbf{r},t), \quad (54)$$

to give the time-dependent intensity of the light emitted by the two atoms, at a detector position where $\varepsilon(\mathbf{r})$ equals unity. Suppose that the two atoms share a single excitation so that their initial state is the superposition

$$|\Psi(t=0)\rangle = [pb_1^{\dagger}(0) + e^{i\phi}\sqrt{1-p^2}b_2^{\dagger}(0)]|0\rangle. \quad (55)$$

Then the expectation value of the intensity operator is

$$\langle I(\mathbf{r},t)\rangle = 2\varepsilon_0 c\{p^2|\mathbf{L}_1|^2 + (1-p^2)|\mathbf{L}_2|^2 + 2p\sqrt{1-p^2}\mathrm{Re}[e^{i\phi}\mathbf{L}_1^{*}\cdot\mathbf{L}_2]\}, \quad (56)$$

where variables $(\mathbf{r},t)$ were dropped.

From Eqs. (53) and (56) it follows that both atoms act as sources and superradiance can take place, even if only the first atom is initially excited (so that $p=1$). This point was stressed for two-atom emission in free space in [49]. The time-dependent intensity that passes at $\mathbf{r}$ is a complicated interference pattern of source fields emitted by four sources: a fast(er) and a (more) slowly decaying source at $\mathbf{R}_1$, and also a fast and a slow source at the atomic position $\mathbf{R}_2$. The photon is shared by and exchanged between the atoms until it is finally emitted, via either the fast superradiant or the slow subradiant decay process. Amplitudes of the fast and slow sources originating from an initially unexcited atom depend both on the interaction between the atoms and on their medium-induced detuning.

VII. APPLICATION: SUPERRADIANCE NEAR A PARTIALLY REFLECTING PLANE

Our multiple-scattering formalism will now be applied to the situation of two identical atomic dipoles in the vicinity of an infinitely thin plane that partially reflects light. We are interested in medium-induced spontaneous-emission rates, Lamb shifts, interatomic interactions, and sub- and superradiant decay rates. These quantities of interest can be expressed in terms of the Green tensor of the medium. In a recent paper [50], we already developed a method to efficiently calculate the Green tensor of a medium consisting of one "plane scatterer" and of several parallel planes, but we only used it to calculate spontaneous-emission rates. For a more detailed discussion of the plane-scatterer model and the method to calculate the Green tensor, we refer to [50]. Below, we give a brief outline of our calculations. We will then focus on those aspects of our results that we believe are generic for many more inhomogeneous dielectrics.

The atomic dipoles are assumed identical and parallel to each other $(\boldsymbol{\mu}_1=\boldsymbol{\mu}_2)$, and parallel to the plane. Moreover, assume the atomic positions $\mathbf{R}_i=(x_i,y_i,z_i)$ to be confined to the line $x_i=y_i=0$. The plane is assumed perpendicular to the z axis. The interatomic interaction Eq. (41) can then be written as

$$J_{12} = J(\mathbf{R}_1,\mathbf{R}_2,\Omega) = \Gamma_0\left(\frac{3c}{4\Omega}\right)\int_0^{\infty} dk_{\parallel}k_{\parallel}(G^{ss}+G^{vv}). \quad (57)$$

Here, G^{ss} stands for the component of the Green tensor $G(k_{\parallel},z_1,z_2,\Omega)$ that describes propagation of s-polarized light, while G^{vv} describes p-polarized light [50]. For a single plane, we have

$$G^{ss}(k_{\parallel},z_1,z_2,\Omega) = G_0^{ss}(k_{\parallel},z_1,z_2,\Omega) + G_0^{ss}(k_{\parallel},z_1,z_{\mathrm{plane}},\Omega) \times T^{ss}(k_{\parallel},\Omega)G_0^{ss}(k_{\parallel},z_{\mathrm{plane}},z_2,\Omega). \quad (58)$$

The free-space tensor component $G_0^{ss}(k_{\parallel},z_1,z_2,\Omega)$ equals $\exp(ik_z|z_1-z_2|)/(2ik_z)$, with the wave vector k_z defined as $\sqrt{(\Omega/c)^2-k_{\parallel}^2}$. The T-matrix $T^{ss}(k_{\parallel},\Omega)$ of the plane for s-polarized light has the form $-[(D_{\mathrm{eff}}(\Omega/c)^2)^{-1}-i/(2k_z)]^{-1}$. The plane is fully characterized by the single parameter D_{eff}, which we call its "effective thickness." We choose the value $D_{\mathrm{eff}}=0.23\lambda$. With this choice, 32% of s-polarized light is transmitted through the plane when averaged over 4π incoming angles. Higher values of D_{eff} give less transmission. For the Green tensor component G^{vv} in Eq. (57) one can write an expression analogous to Eq. (58): in the right-hand side of Eq. (58), the components G_0^{ss} must be replaced by $G_0^{vv}=(k_zc/\Omega)^2G_0^{ss}$, and $T^{ss}(k_{\parallel},\Omega)$ by $T^{vv}(k_{\parallel},\Omega)=-[(D_{\mathrm{eff}}(\Omega/c)^2)^{-1}-ik_zc^2/(2\Omega^2)]^{-1}$.

If in the integral Eq. (57) the in-plane wave vector $k_{\parallel}$ becomes larger than Ω/c, then the wave vector k_z becomes purely imaginary and equal to $i\kappa$, with κ equal to $\sqrt{k_{\parallel}^2-(\Omega/c)^2}$. The semi-infinite integration interval in Eq. (57) therefore falls apart into two parts: a radiative part with $k_{\parallel}$ between 0 and Ω/c, and an evanescent part with $k_{\parallel}$ from Ω/c onwards. The evanescent part of the integral is purely real, except that there is a purely imaginary contribution from a pole in T^{ss} at $\kappa=D_{\mathrm{eff}}(\Omega/c)^2/2$. This pole corresponds to an s-polarized guided mode. Near the pole, the real evanescent part of the integral over the s-wave integrand in Eq. (57) must be taken as a Cauchy principal-value integral. There is no corresponding pole in T^{vv}. The evanescent part of the integral for p-polarized light is purely real and can be evaluated numerically right away. All relative errors in our numerical results are smaller than 10^{-6}.

The single-atom spontaneous-decay rate Eq. (31) can be found from the interaction Eq. (41) through the relation $\Gamma(\mathbf{R}_1,\Omega)=-2\mathrm{Im}\,J_{12}(\mathbf{R}_1,\mathbf{R}_1,\Omega)$, while the the Lamb shift Eq. (32) follows from $\Delta(\mathbf{R}_1,\Omega)=\mathrm{Re}[J_{12}(\mathbf{R}_1,\mathbf{R}_1,\Omega)$

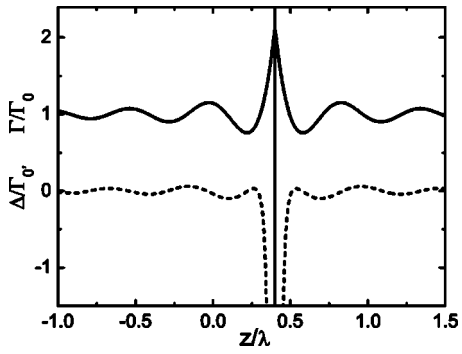

FIG. 1. Spontaneous-emission rate Γ (solid line) and Lamb shift Δ (dashed line) as a function of the position of an atom near a partially reflecting plane. The effective thickness of the plane is $D_{\mathrm{eff}}=0.23\lambda$. The atomic dipole moment points parallel to the plane. The plane is positioned at $z_{\mathrm{plane}}=0.4\lambda$, to make comparisons with later figures easier. Both Γ and Δ are given in units of Γ_0, and the positions z are scaled to the wavelength $\lambda=2\pi c/\Omega$ of the emitted light. The period of the damped oscillations in both Γ and Δ is $\lambda/2$.

$-J_{12}^{(0)}(\mathbf{R}_1,\mathbf{R}_1,\Omega)]$. Figure 1 shows how single-atom properties are modified by the presence of the plane. The figure shows a peak in the decay rate near the plane due to emission into the guided modes [50]. Away from the plane, the decay rate shows damped oscillations towards the free-space decay rate Γ_0. There are two oscillations per wavelength λ, a characteristic also well-known for spontaneous emission near a perfect mirror [44]. The Lamb shift shows similar damped oscillations around $\Delta=0$ away from the plane. At distances less than $\lambda/10$ the shift becomes strongly negative and it actually diverges to minus infinity. The atom is attracted to the plane [44], but here we assume atomic positions to be fixed.

In Fig. 2 we present dipole-dipole interactions for two atoms near a plane. The first atom is kept fixed in the origin,

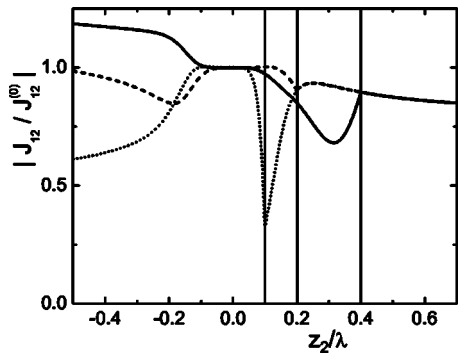

FIG. 2. Absolute values of interatomic interactions $J_{12}(\mathbf{R}_1,\mathbf{R}_2,\Omega)$ near a partially reflecting plane, scaled to the free-space interaction strength $|J_{12}^{(0)}(\mathbf{R}_1,\mathbf{R}_2,\Omega)|$. The plane is as in Fig. 1. The first atom's position is fixed in the origin. The second atom travels along the line $(x_2=y_2=0,z_2)$. The atomic dipoles point in the same directions, parallel to the plane. The three graphs differ in the position of the plane with respect to the first atom. Solid line: $z_{\mathrm{plane}}/\lambda=0.4$; dashed line: $z_{\mathrm{plane}}/\lambda=0.2$; dotted line: $z_{\mathrm{plane}}/\lambda=0.1$. (All three planes are shown, but in each case considered only a single plane is present.)

the distance of the plane to this first atom is chosen, and then the absolute value of the interatomic interaction J_{12} is plotted as a function of the position of the second atom, relative to the free-space value $|J_{12}^{(0)}|$. The interaction is the sum of radiative and evanescent interactions, of both *s*-polarized and *p*-polarized light. The figure shows that for z_2 approaching $z_1=0$, the relative difference between $|J_{12}|$ and the (divergent) free-space interaction strength $|J_{12}^{(0)}|$ becomes negligible, irrespective of the position of the plane. This holds independently of the reflectivity of the plane (not shown in Fig. 1). Interestingly, the dipole-dipole interaction is also independent of the position of the plane (but not of its reflectivity) if the plane stands in between the two atoms. In other words, with the atomic positions fixed at either side of the plane, one can move the plane back and forth without changing the interatomic interaction. This fact can be read off from Fig. 2 for $z_2/\lambda>0.4$, where the three graphs (corresponding to three plane positions) overlap. It can also be understood from the form of the interaction in Eq. (57), because all terms in the interaction either depend on $|z_2-z_1|$ or on $(|z_1-z_{\mathrm{plane}}|+|z_2-z_{\mathrm{plane}}|)$. For $z_2<0$ and $|z_2|\gg\lambda$, the relative interaction $|J_{12}/J_{12}^{(0)}|$ approaches a constant value, which can be either larger or smaller than unity, depending on the distance of the plane to atom 1. As a check on our calculations (not shown), we found that interatomic interactions vanished (as expected) when an almost ideal mirror (a plane with $D_{\mathrm{eff}}=100\lambda$) is placed in between them.

Figure 3(a) shows two-atom superradiant and subradiant decay rates, as modified by the presence of the plane. The plots are based on Eqs. (48) and (57). The complex square root in Eq. (48) has solutions that differ by an overall minus sign. Care was taken to choose the solution from the same branch as we varied the position of the second atom. Without the plane, one would have had $\Gamma_{0,\pm}=\Gamma_0\mp 2\ \mathrm{Im}\ J_{12}^{(0)}$. With the plane, the medium-induced detuning becomes negligible as z_2 approaches z_1. Then Γ_-/Γ_0 vanishes, as in free space. The corresponding small-distance limit of Γ_+/Γ_0 is not equal to 2 as for free space, but rather twice the single-atom decay rate $\Gamma=1.14\Gamma_0$ in the presence of the plane. If the second atom moves towards the mirror, then the medium-induced detuning (see Fig. 1) grows fast while the dipole-dipole interaction (Fig. 2) decreases. With Eq. (48) we then find that $\Gamma_-\simeq\Gamma_2$ and $\Gamma_+\simeq\Gamma_1$. Indeed, for z_2 closer than $\lambda/10$ to the plane, Γ_- follows the single-plane emission rate of Fig. 1, while Γ_+ equals $\Gamma(\mathbf{R}_1,\Omega)=1.14\Gamma_0$. With atom 2 so close to the plane, superradiance is completely absent, even though the identical atoms are less than half a wavelength apart. For $z_2>0.5\lambda$ or $z_2<0.3\lambda$, detuning has become less important and the decay rates follow (not quite sinusoidal) damped oscillations. Their period is λ, as it is for superradiance in free space.

Figure 3(b) again shows super- and subradiant decay rates, now also for larger interatomic distances. For $-4\lesssim z_2/\lambda\lesssim 3$, the rates $\Gamma_\pm$ exhibit the same damped oscillations with period λ that we also saw in Fig. 3(a). However, for z_2/λ smaller than -4 or larger than 3, $\Gamma_\pm$ show *two* oscillations per wavelength, like we saw for the single-atom decay rate in Fig. 1. Hence we can identify a rather sharp crossover regime at a few wavelengths away from the plane between superradiance and single-atom emission. For larger dis-

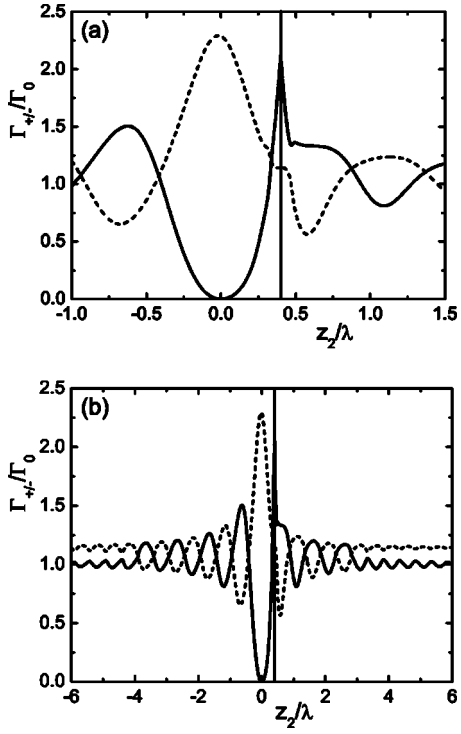

FIG. 3. Subradiant and superradiant decay rates for two atoms near a partially reflecting plane, as a function of the position of the second atom. The situation is as in Fig. 2 with the plane fixed at $z_{\text{plane}}/\lambda = 0.4$. Figure (a) zooms in around the plane, showing a vanishing subradiant decay rate Γ_- (solid line) as the second atom approaches the first one in the origin. The superradiant decay rate Γ_+ (dashed line) becomes more than twice the single-atom decay rate Γ_0. Both Γ_- and Γ_+ show perturbed oscillations on the scale of λ. Figure (b) zooms out to larger distances, showing a crossover regime between damped oscillations with a period λ and more distant oscillations with a period $\lambda/2$.

tances, again medium-induced detuning dominates the dipole-dipole interaction. Indeed for large distances we see the same behavior as very close to the plane, namely that Γ_- approaches Γ_2 (which at these positions almost equals Γ_0) while Γ_+ has the limiting value $\Gamma_1 = 1.14\Gamma_0$. In the crossover regime, $|J_{12}|$ has the same order of magnitude as the detuning $|\Delta_1 - \Delta_2 - i(\Gamma_1 - \Gamma_2)/2|$.

If one puts the plane closer to the first atom, then this atom becomes further detuned from its free-space properties. The crossover should then take place with the second atom at shorter distances where the interaction is still stronger. This we have verified (not shown). At a fixed frequency, the spatial intervals in which superradiance occurs therefore depend on three distances, namely the interatomic distance and the distances between each atom and the plane.

Not only the super- and the subradiant emission rates are influenced by the presence of the plane, but also the source amplitudes of the two atoms are modified, shown in Eq. (53): if initially only the first atom is excited, then the source amplitude of the second atom is modified by a factor $C_{1\pm} \equiv 1 \pm \sin\alpha$, for superradiant (+) and subradiant (−) decay, respectively. Atom 2 gets a factor $C_2 \equiv \cos\alpha$ for both decay processes. Figure 4 shows $|C_2/C_{1+}|$ as z_2 is varied. The emis-

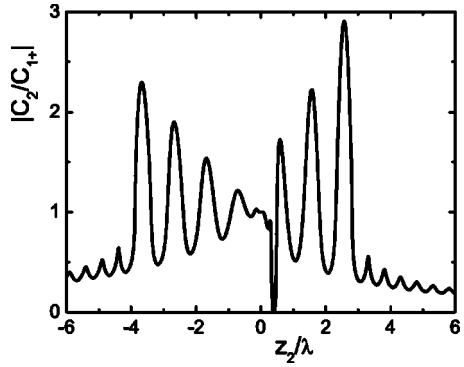

FIG. 4. Absolute value of the source amplitude $C_2 = \cos\alpha$ of the second atom, divided by the source amplitude $C_{1+} = (1 + \sin\alpha)$ of the first atom. The situation is as in Fig. 3. The amplitudes are associated with superradiant emission; see Eq. (53).

sion rates of Fig. 3 and the relative amplitudes in Fig. 4 show the same crossover regions between oscillations with periods $\lambda/2$ and λ. When the two atoms coincide, the detuning vanishes and $|C_2/C_{1+}|$ equals unity. (For free space, $|C_2/C_{1+}|$ equals unity everywhere, even if the atoms do not coincide.) The coinciding atoms are equivalent and superradiance can occur. On the other hand, close to the plane at $|z_2/\lambda - 0.4| \lesssim 0.1$, atom 2 is strongly detuned and $|C_2/C_{1+}|$ vanishes: the second atom emits none of the light initially residing in the first one and superradiance does not occur.

Figure 4 also shows that at larger distances ($z_2 \lesssim -4$ or $z_2 \gtrsim 3$), detuning is again strong enough to make emission by the second atom less probable than in free space. At these larger distances, the medium-induced detuning suppresses the net transfer of light from atom 1 to atom 2 and superradiance does not occur. In the two intervals $-4 < z_2\lambda < 0.3$ and $0.5 < z_2/\lambda < 3$ where superradiance does occur, we see that the peaks of the relative source amplitude $|C_2/C_{1+}|$ are higher than unity. There the probability that light initially residing on the first atom is finally emitted by the second one is higher than in free space. The peaks of Fig. 4 correspond to positions of the second atom for which most light is finally emitted by the second atom, although initially only the first atom was excited. Interestingly, the peaks of $|C_2/C_{1+}|$ become higher as the second atom moves away from the first. The highest peaks occur when the complex-valued dipole-dipole interaction (almost) exactly compensates the complex-valued detuning. (Such a resonant situation does not exist for identical atoms in free space.) For larger $|z_2|$, the dipole-dipole interaction becomes too weak to compensate for the detuning and Fig. 4 shows very abrupt transitions from superradiance to single-atom emission on both sides of the plane.

VIII. CONCLUSIONS, DISCUSSION, AND OUTLOOK

In this paper, a multiple-scattering theory was set up with at its heart the Lippmann-Schwinger equation (11) that describes the electromagnetic field operators in an inhomogeneous dielectric with guest atoms present. We solved the LS equation exactly in terms of the properties of the atoms and

the Green tensors of the medium, both when one and when several guest atoms are present. The solution for the electric field operator has three parts: a part that has not seen the guest atoms, a part that describes the scattering by the resonant atoms, and a part that describes the atoms as sources.

Our formalism is a generalization of an already existing point-scattering formalism for classical waves. The generalization is twofold: first, our formalism is valid not only for free space but for atoms in all dielectrics that can be described macroscopically in terms of a real relative dielectric constant $\varepsilon(\mathbf{r})$. Second, it is a multiple-scattering theory in quantum optics rather than classical optics. In relation to this point we find the double nature of atoms both as scatterers and as sources of light. The formalism is quantum mechanical in the sense that it can describe the propagation and scattering of nonclassical sources of light. These can be either external or atomic sources. In quantum optics, the medium must be described with more care, just like the quantum and classical descriptions of a beam splitter differ [18]. As for the beam splitter, classical light sources give classical measured fields in our formalism, since we described the guest atoms as harmonic oscillators.

A nice feature of the LS equation (11) is that it follows exactly from a dipole Hamiltonian that is the result of a canonical quantization theory. The Hamiltonian describes guest atoms microscopically and treats the dielectric macroscopically. The atomic dipoles do not couple to the electric field operator $\mathbf{E}$ but rather to a field operator that we call $\mathbf{F}$ and that includes the atom's own polarization field. For free space this is a well known result. We find that the propagator for the field $\mathbf{F}$ in our LS equation is not the ordinary Green tensor G, but rather a Green tensor that we called K. There exists a simple relation (22) between G and K for an arbitrary dielectric. G can be split into the generalized transverse Green tensor G^T that propagates the vector potential $\mathbf{A}$, and the longitudinal Green tensor G^L.

In the Appendix we showed that the volume-integrated electric field (A6) produced in free space by an atomic dipole is equal to minus one third of its polarization field. This is an operator relation at finite frequency. A different (incorrect) relation would have resulted if the field $\mathbf{F}$ had been interpreted as the electric-field operator. We have not come across other work that addresses the relation between the dipole interaction, the occurrence of K rather than G in a multiple-scattering theory, and the volume-integrated electric field around a dipole. In this respect, our formalism also sheds new light on quantum optics in free space.

The infinitely sharp single-atom resonance in the potential V obtains a radiative shift and a width in the T-matrix T. In our formalism, the position-dependent shift and decay rate are the summed effects of infinitely many light-scattering events off the atomic potential. The scattered-field operator for a single atom contains two parts: an elastic-scattering term and a term describing resonance fluorescence. Direct interatomic interactions are absent in the dipole Hamiltonian (1c). Dipole-dipole interactions appear "dynamically" in the solutions of the Lippmann-Schwinger equation for several atoms. An inhomogeneous medium modifies both the longitudinal and the generalized transverse dipole-dipole interactions; see Eqs. (42a) and (42b).

The multiple-scattering formalism has been used to study superradiance in an inhomogeneous medium. The often dominant electronic component to inhomogeneous broadening was neglected in order to focus on photonic effects. As an application, we studied how dipole-dipole interactions and two-atom superradiance are influenced by a partially reflecting plane. We found position-dependent modifications of dipole-dipole interactions. For our choice of parameters, the plane suppresses superradiance if one of the atoms is very close or very far from the plane. Both atoms will then emit as if alone. For intermediate distances, two-atom sub- and superradiance will occur. Due to the plane, emission rates are modified and so are the relative amplitudes of the atomic sources. Interestingly, we found that medium-induced complex detuning can lead to enhanced transfer of light from the one atom to the other, before superradiant emission occurs. Also, we found sharp crossovers between spatial intervals where superradiance occurs (with decay rates oscillating once per wavelength) and single-atom emission (two oscillations per wavelength).

The length of the intervals in which superradiance occurs depends on the atomic positions with respect to each other and to the plane. This length could be called a "perpendicular coherence length." This would complement the concept of a transverse coherence length (or effective mode radius) [20,51]. The latter concept is used in the analysis of cooperative emission in a planar microcavity when the atoms have the same z-coordinate, but have different coordinates in another direction. An important difference between the two lengths is that only the perpendicular coherence length is influenced by medium-induced detuning.

We believe that our results for cooperative emission near the plane are generic and that similar crossover regions will occur in more complex dielectrics. Still, it would be interesting to study the influence of other dielectric structures on multiatom processes, Bragg mirrors for example, or "optical corrals" [52,53]. Photonic crystals are also very interesting media, for which superradiance has only been studied in an isotropic model [21–23] where all position dependence is neglected. Like near a plane, superradiance inside a real photonic crystal will be influenced by medium-induced detuning. As another application of our formalism, statistical distributions of optical proximity resonances of many-atom systems can be studied, to find analogies and differences in inhomogeneous optical and electronic systems [54].

We made a pole approximation in a late stage of our formalism, after which we found exponential atomic decay. The pole approximation no longer holds when the atom-field interaction becomes strong [55]. The approximation also breaks down if local densities of states jump steeply as a function of frequency near the atomic transition frequency. There is a current debate whether pole approximations will break down at the band edges of realistic three-dimensional photonic crystals [56], like it is found for the isotropic model [57]. In principle, our formalism could also be used without making the pole approximation.

Our theory is valid if frequency dispersion of the medium can be neglected. Now single-atom emission rates only depend on one frequency of the medium, so that dispersion is not important. On the other hand, radiative line shifts, inter-

atomic interactions, and hence superradiant decay rates do depend on all frequencies of the medium. In our example of two atoms near a plane, the immediate vicinity of the atoms was free space. However, for atoms embedded in a $\varepsilon \neq 1$ part of a medium, line shifts would diverge unless frequency dispersion of the medium is taken into account [58]. This will also be the case for position-dependent radiative shifts in photonic crystals [59]. It will be interesting to study the influence of frequency dispersion of the medium on cooperative atomic emission, for example based on Refs. [31,60].

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APPENDIX: HOMOGENEOUS DIELECTRIC

1. Delta and Green functions

For a homogeneous dielectric with refractive index n, the property "generalized transverse" reduces to transverse in the ordinary sense. The medium is translational invariant, so that $\boldsymbol{\delta}^{T,L}(\mathbf{r},\mathbf{r}') = \boldsymbol{\delta}^{T,L}(\mathbf{r}-\mathbf{r}')$. The transverse and longitudinal delta functions appearing in Eqs. (17) and (19b) now become

$$\boldsymbol{\delta}^{T}_{\mathrm{hom}}(\mathbf{r}) = \frac{2}{3}\delta(\mathbf{r})\mathbf{I} - \frac{1}{4\pi r^3}(\mathbf{I} - 3\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}), \tag{A1a}$$

$$\boldsymbol{\delta}^{L}_{\mathrm{hom}}(\mathbf{r}) = \frac{1}{3}\delta(\mathbf{r})\mathbf{I} + \frac{1}{4\pi r^3}(\mathbf{I} - 3\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}), \tag{A1b}$$

where $\hat{\mathbf{r}}$ is defined as $\mathbf{r}/|\mathbf{r}|$, the unit vector in the direction of $\mathbf{r}$. The sum of the transverse and the longitudinal delta function is simply $\delta(\mathbf{r})\mathbf{I}$, since their "dipole" parts cancel. Notice that n does not enter these delta functions. The derivation follows the free-space treatment [48].

The dyadic Green function $\mathbf{G}_{\mathrm{hom}}(\mathbf{r},\mathbf{r}') = \mathbf{G}_{\mathrm{hom}}(\mathbf{r}-\mathbf{r}')$ for the homogeneous medium is the sum of a transverse and a longitudinal part. The transverse part is [40]

$$\mathbf{G}^{T}_{\mathrm{hom}}(\mathbf{r},\omega) = -\frac{\mathbf{I} - 3\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}{4\pi(n\omega/c)^2 r^3} - \frac{e^{in\omega r/c}}{4\pi r}[P(in\omega r/c)\mathbf{I} + Q(in\omega r/c)\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}], \tag{A2a}$$

with the function $P(z)$ defined as $(1-z^{-1}+z^{-2})$ and $Q(z)$ as $(-1+3z^{-1}-3z^{-2})$. With the use of the definition (19a) of the longitudinal Green function and the transverse delta function (A1a), the longitudinal Green function is found to be

$$\mathbf{G}^{L}_{\mathrm{hom}}(\mathbf{r},\omega) = \frac{\mathbf{I} - 3\hat{\mathbf{r}}\otimes\hat{\mathbf{r}}}{4\pi(n\omega/c)^2 r^3} + \frac{\delta(\mathbf{r})}{3(n\omega/c)^2}\mathbf{I}. \tag{A2b}$$

The delta-function term in $\mathbf{G}^{L}_{\mathrm{hom}}$ appears naturally and there was no need to add it "by hand" as is done elsewhere [40,61].

2. Volume-integrated dipole field

The rigorous multiple-scattering formalism of Sec. II with the Green functions $\mathbf{K}$ will now be used to calculate the volume integral of the electric-field operator $\mathbf{E}$ in terms of the atomic polarization fields $\Sigma_m \mathbf{P}_m$ of Eq. (3), with the volume taken over a small sphere enclosing an atom.

With the help of the Eqs. (9a) and (9b), and the definitions of the source fields (12) and potentials (13), the polarization field in frequency space can be related to other operators as

$$\mathbf{P}_m(\omega) = -\left(\frac{\varepsilon_0 c^2}{\omega^2}\right)[\mathbf{S}_m(\omega) + \mathbf{V}_m(\omega)\cdot\mathbf{F}(\mathbf{R}_m,\omega)]. \tag{A3}$$

There exists therefore a simple relationship between the field $\mathbf{F}$ and the polarization fields [use Eq. (11)]

$$\mathbf{F}(\mathbf{r},\omega) = \mathbf{E}^{(0)}(\mathbf{r},\omega) - \frac{\omega^2}{\varepsilon_0 c^2}\sum_{m=1}^{N}\mathbf{K}(\mathbf{r},\mathbf{R}_m,\omega)\cdot\mathbf{P}_m(\omega). \tag{A4}$$

This equation is still valid for all inhomogeneous dielectrics. Now assume that the sources are in free space. Consider the volume-integral of the field $\mathbf{F}$ over a small sphere (denoted by $\odot$) containing only the source at $\mathbf{R}_m$, at its center. The integral is determined by the free-space Green function $\mathbf{K}_0(\mathbf{r},\mathbf{R}_m,\omega)$ for positions $\mathbf{r}$ close to $\mathbf{R}_m$ [see Eqs. (21) and (22)]. The transverse Green function $\mathbf{G}^{T}_0(\mathbf{r}-\mathbf{R}_m,\omega)$ in $\mathbf{K}_0$ has a vanishing contribution to the integral, since its pole goes as $|\mathbf{r}-\mathbf{R}_m|$ at short distances [see Eq.(A2a)]. The dipole part of the transverse delta function (A1a) has a vanishing angle-integral over the sphere and does not contribute either. What remains is the delta-function part of the transverse delta function, which gives the radius-independent result

$$\int_{\odot} d\mathbf{r}\ \mathbf{F}(\mathbf{r},\omega) = \frac{2}{3\varepsilon_0}\mathbf{P}_m(\omega). \tag{A5}$$

Now the subtlety becomes important that the field $\mathbf{F} = -\mathbf{D}(\mathbf{R})/[\varepsilon_0\varepsilon(\mathbf{R})]$ is equal to the electric field $\mathbf{E}$ everywhere except at the positions of the guest atoms [see Eq. (4)]. The expression in Eq. (A5) is therefore not equal to the volume-integrated electric field. With the definitions of the fields $\mathbf{D}$ and $\mathbf{F}$ given in Sec. II A, one obtains the relation

$$\int_{\odot} d\mathbf{r}\ \mathbf{E}(\mathbf{r},\omega) = -\frac{1}{3\varepsilon_0}\mathbf{P}_m(\omega). \tag{A6}$$

The static and classical version of this "sum rule" is presented for example in [61]. There, and more recently in [40], a delta function is added by hand to the static dipole field or to the longitudinal Green function. In contrast, Eq. (A6) was found here as an operator relation without adding any terms by hand.

The interpretation of the field to which a dipole couples is not just a matter of choice in the present formalism. If one wrongly identifies $\mathbf{F}$ as the electric field $\mathbf{E}$ but correctly derives the relation (21) or (22) between $\mathbf{K}$ and $\mathbf{G}$, then the wrong volume-integrated electric field $2\mathbf{P}_m(\omega)/(3\varepsilon_0)$ would have resulted. The delta function term that is the difference between $\mathbf{G}$ and $\mathbf{K}$ in Eq. (22) and the difference between the

field operators **E** and **F** have the same physical origin: the atomic polarization field.

Still, there is nothing truly quantum mechanical about the sum rule (A6). In a classical canonical theory, one would find the same dipole coupling $-\boldsymbol{\mu}\cdot\mathbf{F}$ and Green function **K**. However, a canonical formalism is usually by-passed in classical optics. It is then assumed that a classical dipole couples to the classical electric field and furthermore that light propagates from a source according to the Green function **G** rather than **K**. By summing Eqs. (A2a) and (A2b) for $n=1$, one finds that $\mathbf{G}_0$ naturally has the correct delta-function term to produce Eq. (A6). Therefore, although following a less rigorous procedure, one has the luck that there is no need to add terms by hand in order to derive Eq. (A6) classically.

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