Radiative atom-atom interactions in optically dense media: Quantum corrections to the Lorentz-Lorenz formula

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Generalized single-atom Maxwell-Bloch equations for optically dense media are derived taking into account non-cooperative radiative atom-atom interactions. Applying a Gaussian approximation and formally eliminating the degrees of freedom of the quantized radiation field and of all but a probe atom leads to an effective time-evolution operator for the probe atom. The mean coherent amplitude of the local field seen by the atom is shown to be given by the classical Lorentz-Lorenz relation. The second-order correlations of the field lead to terms that describe relaxation or pump processes and level shifts due to multiple scattering or reabsorption of spontaneously emitted photons. In the Markov limit a nonlinear and nonlocal single-atom density matrix equation is derived. To illustrate the effects of the quantum corrections we discuss amplified spontaneous emission and radiation trapping in a dense ensemble of initially inverted two-level atoms and the effects of radiative interactions on intrinsic optical bistability in coherently driven systems.

I. INTRODUCTION

The interaction of the radiation field with a dilute ensemble of atoms is usually described in the semiclassical and dipole approximation by the well-known Maxwell-Bloch equations. This description fails to be accurate, however, when a dense medium is considered.

Since the early work of H. A. Lorentz and L. Lorenz [1] it is known that the classical local field, that couples to an atom in a dense medium, differs from the macroscopic (Maxwell) field by a term proportional to the medium polarization [2,3]. The most prominent effects of the Lorentz-Lorenz (LL)-correction in dense media are the change of the linear index of refraction according to the Clausius-Mossotti relation [2], the enhancement of nonlinear susceptibilitiesΦs [4], shifts and deformation of resonance lines [5,6], intrinsic optical bistability [7,8], and piezo-photonic switching [9].

On the other hand the quantum nature of the radiative atom-atom interaction can drastically influence the behavior of the ensemble. In the extreme case of anisotropic, high-density samples, excited atoms can cooperatively emit spontaneous photons, a phenomenon known as superradiance [10–12]. But even if the system does not fulfill the conditions for cooperative evolution, the presence of spontaneous photons and the associated effects like amplified spontaneous emission (or superluminescence) and radiation trapping [13] can not be neglected. Imprisonment of incoherent photons especially affects otherwise long-lived ground-state coherences. We therefore expect radiative atom-atom interactions to be important in areas such as resonant linear and nonlinear optics based on atomic phase coherence [14,15], cooling of atoms and Bose-Einstein condensation via velocity-selective coherent population trapping [16] and optical computing.

Another important effect of large atomic densities is the increase of atomic collisions. Here we will not consider these effects, however, and focus our attention entirely on radiative interactions.

In the present paper we study the atomic evolution in a dense medium irradiated by external coherent light fields. The macroscopic classical radiation field in the medium obeys Maxwell's equations with the mean atomic polarization as source term. To derive equations of motion for the many-atom system, we start from a nonrelativistic quantized interaction Hamiltonian. Thus interactions between the atoms mediated by the quantized radiation field such as reabsorption and scattering of spontaneous photons are taken into account.

Our aim is to derive an effective single-atom density-matrix equation. For this we introduce an interaction picture with the radiation field coupling to all other atoms. Assuming a Gaussian (and therefore classical) statistics of the interacting field, we can formally eliminate its degrees of freedom from the probe-atom time evolution. In the Markov limit of short-lived field correlations this yields a density-matrix equation for the probe atom. We will show that the mean coherent amplitude seen by the probe atom differs from the macroscopic Maxwell field by a term proportional to the mean polarization of the medium in agreement with the classical Lorentz-Lorenz relation [2]. In addition, the density matrix equation contains relaxation and level-shift terms, which describe reabsorbing and multiple scattering of spontaneously emitted photons. The corresponding relaxation rates and frequency shifts are proportional to the spectrum of the incoherent part of the radiation inside the medium. This spectrum is also the Fourier-transform of a certain 2-time Greensfunction, for which we derive a Dyson equation. A formal solution of the Dyson equation allows to express the incoherent spectrum in terms of atomic variables. Thus we eventually obtain a closed, *nonlinear* and spatially *nonlocal* density matrix equation of Lindblad-type.

Our paper is organized as follows. In Sec. II we derive the effective single-particle time-evolution operator by formally eliminating the degrees of freedom of the quantized radiation field interacting with the background atoms. In Gaussian approximation this operator contains first and second-order field cumulants. In the Markov limit of spectrally broad field correlations, a density matrix equation is obtained. In Sec. III we show that the first-order term leads to the Lorentz-Lorenz relation between the coherent amplitude of the local field, the mean field amplitude in the medium (Maxwell field), and the mean polarization. In Sec. IV we derive a Dyson equation for the second-order field cumulants or 2-point Greens-functions and formally solve them in terms of single-atom density matrix elements. The resulting nonlinear density matrix equation is discussed in Sec.V for the examples of amplified spontaneous emission and radiation trapping in an inhomogeneously broadened system of initially excited two-level atoms and intrinsic optical bistability in a strongly driven dense ensemble of two-level atoms.

II. EFFECTIVE TIME-EVOLUTION OF ATOMS

A. Formal elimination of the quantized radiation field

We here consider an ensemble of atoms interacting with the quantized radiation field under conditions which justify the dipole and rotating-wave approximation (RWA). Since we are interested in the dynamics of a single atom, we distinguish a probe atom at position $\vec{r}_0$ with a dipole operator $\vec{p}$ and environment atoms at positions $\vec{r}_j$ whose dipole operators are denoted by $\vec{d}^j$. The Hamiltonian of the system is given by

$$H = \sum_j H_0^j + H_{\rm field} - \vec{p}\cdot\Big[\vec{E}(\vec{r}_0) + \vec{\mathcal{E}}(\vec{r}_0)\Big] - \sum_{j\neq 0} \vec{d}^{\,j}\cdot\Big[\vec{E}(\vec{r}_j) + \vec{\mathcal{E}}(\vec{r}_j)\Big], \qquad (1)$$

where H_0^j and $H_{\rm field}$ are the free Hamiltonians of the jth atom and the quantized radiation field respectively, and we have split the field in an operator component E and an external classical driving field $\mathcal{E}$. We use an interaction picture where the time evolution is described by

$$S = T\exp\left\{-\frac{i}{\hbar}\int_{-\infty}^{\infty} d\tau\;\, V_{\rm p}(\tau)\right\} = T\exp\left\{\frac{i}{\hbar}\int_{-\infty}^{\infty} d\tau\;\, p(\tau)\left[E(\vec{r}_0,\tau)+\mathcal{E}(\vec{r}_0,\tau)\right]\right\}, \qquad (2)$$

were T denotes time-ordering and the field operator E still contains the coupling to all other atoms. For notational simplicity we have suppressed vector indices of the dipole moment and electric field. With the help of (2) any (time-ordered) correlation function of probe-atom operators A_H and B_H in the Heisenberg-picture (subscript "H") can be related to interaction picture operators via

$$\langle T^{-1}[A_H(t_1)A_H(t_2)]T[B_H(t_3)B_H(t_4)]\rangle = \Big\langle T^{-1}[S^{-1}A(t_1)A(t_2)]T[SB(t_3)B(t_4)]\Big\rangle\,, \qquad (3)$$

where $\langle\cdots\rangle$ stands for $Tr\{\rho_0\cdots\}$ with $\rho_0=\rho(-\infty)$ being the initial density operator at $t=-\infty$.

A very helpful formal simplification of Eq.(3) can be achieved by introducing the so-called Schwinger-Keldysh time contour C [17] shown in Fig. 1 which starts at $t=-\infty$, goes to $t=+\infty$ and back $t=-\infty$. Each physical time correspond two times on the contour. A time ordering operator T_C is introduced, which is identical to T on the upper branch $(+)$ and to T^{-1} on the lower branch $(-)$ of the contour and orders all operators with time arguments on $(-)$ to the left of those with time arguments on $(+)$.

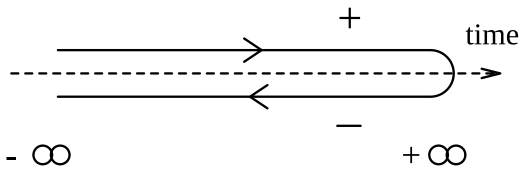

FIG. 1. Schwinger-Keldysh time contour

With these definitions we can write Eq.(3) with a single exponential time-evolution operator. This will considerably simplify the following elimination procedure.

$$\langle[T^{-1}A_H(t_1)A_H(t_2)][TB_H(t_3)B_H(t_4)]\rangle = \langle T_C[S_C A(t_1^-)A(t_2^-)B(t_3^+)B(t_4^+)]\rangle, \qquad (4)$$

where the superscripts $\pm$ specify the branch of the contour, and

$$S_C = T_C\exp\left\{-\frac{i}{\hbar}\int_C d\check{\tau}\;\, V_{\rm p}(\check{\tau})\right\}, \qquad (5)$$

with $\check{\tau}$ denoting a time on C.

We now formally eliminate the degrees of freedom of the quantized radiation field and the environment atoms by tracing over the corresponding states. In order to express the expectation value of an exponential operator again as an exponential operator, i.e. as a new – effective – time-evolution operator, we use a generalization of the cumulant generating function for a classical stochastic variable X [18,19]:

$$\Big\langle\exp\{sX\}\Big\rangle_X = \exp\left\{\sum_{m=0}^{\infty}\frac{s^m}{m!}\langle\langle X^m\rangle\rangle\right\}, \qquad (6)$$

where the $\langle\langle X^m\rangle\rangle$ are the cumulants, which have the following explicit form

$$\langle\langle X\rangle\rangle = \langle X\rangle, \qquad (7)$$

$$\langle\langle XY\rangle\rangle = \langle XY\rangle - \langle X\rangle\langle Y\rangle, \quad \text{etc.} \qquad (8)$$

As can bee seen from (6), the elimination procedure leads in general to an infinite number of terms in the effective action. To make the problem tractable, we will however assume that the radiation field is Gaussian, i.e. that all cumulants $\langle\langle E^m\rangle\rangle$ with $m>2$ vanish identically. This is a consistent and for our purposes well justified approximation. With this we find

$$S_C^{\rm eff} = \langle S_C\rangle_{\rm field} \tag{9}$$
$$T_C \exp\left\{\frac{i}{\hbar}\int_C d\check\tau\, p(\check\tau)\Big[\mathcal{E}(\vec r_0,\check\tau)+\langle E(\vec r_0,\check\tau)\rangle\Big]\right.$$
$$\left.-\frac{1}{2\hbar^2}\int_C d\check\tau_1\int_C d\check\tau_2\, p(\check\tau_1)\,\mathcal{D}(\vec r_0,\check\tau_1;\vec r_0,\check\tau_2)\,p(\check\tau_2)\right\},$$

where

$$\mathcal{D}_{\mu\nu}(\check 1,\check 2)=\Big\langle\Big\langle T_C\, E_\mu(\vec r_1,\check\tau_1)E_\nu(\vec r_2,\check\tau_2)\Big\rangle\Big\rangle \tag{10}$$

is a (tensorial) Greensfunction (GF) of the interacting electric field, and we have used the abbreviations $\check 1\equiv\vec r_1,\tau_1$ and $\check 2\equiv\vec r_2,\tau_2$. Note, that we used a short notation, and $p(\check 1)\,\mathcal{D}(\check 1,\check 2)\,p(\check 2)$ in Eq.(9) should read $\sum_{\alpha,\beta=1}^3 p_\alpha(\check 1)\,\mathcal{D}_{\alpha\beta}(\check 1,\check 2)\,p_\beta(\check 1,\check 2)$. We now apply the rotating-wave approximation. For this we introduce slowly varying positive and negative frequency components,

$$p(\check\tau)=p^+(\check\tau)+p^-(\check\tau)=\widetilde p^{\,+}(\check\tau)e^{-i\omega\tau}+\widetilde p^{\,-}(\check\tau)e^{i\omega\tau}, \tag{11}$$
$$E(\check\tau)=E^+(\check\tau)+E^-(\check\tau)=\widetilde E^+(\check\tau)e^{-i\omega\tau}+\widetilde E^-(\check\tau)e^{i\omega\tau}, \tag{12}$$

with ω being the transition frequency of the considered probe atom, and neglect combinations of the type p^+E^+ and p^-E^-. Thus we have

$$S_C^{\rm eff} =$$
$$T_C \exp\left\{\frac{i}{\hbar}\int_C d\check\tau\Big[p^+(\check\tau)\mathcal{E}_L^-(\vec r_0,\check\tau)+p^-(\check\tau)\mathcal{E}_L^+(\vec r_0,\check\tau)\Big]\right.$$
$$-\frac{1}{2\hbar^2}\int_C d\check\tau_1\int_C d\check\tau_2\Big[\widetilde p^{\,+}(\check\tau_1)\,D(\vec r_0,\check\tau_1;\vec r_0,\check\tau_2)\,\widetilde p^{\,-}(\check\tau_2)$$
$$\left.+\widetilde p^{\,-}(\check\tau_1)\,C(\vec r_0,\check\tau_1;\vec r_0,\check\tau_2)\,\widetilde p^{\,+}(\check\tau_2)\Big]\right\}, \tag{13}$$

where

$$\mathcal{E}_{L\mu}(\vec r,t)=\mathcal{E}_\mu(\vec r,t)+\langle E_\mu(\vec r,t)\rangle \tag{14}$$

is the local field seen by the probe atom, and

$$D_{\mu\nu}(\vec r_0,\check\tau_1;\vec r_0,\check\tau_2) = \langle\langle T_C E_\mu^-(\vec r_0,\check\tau_1)E_\nu^+(\vec r_0,\check\tau_2)\rangle\rangle\, e^{-i\omega(\tau_1-\tau_2)}, \tag{15}$$
$$C_{\mu\nu}(\vec r_0,\check\tau_1;\vec r_0,\check\tau_2) = \langle\langle T_C E_\mu^+(\vec r_0,\check\tau_1)E_\nu^-(\vec r_0,\check\tau_2)\rangle\rangle\, e^{+i\omega(\tau_1-\tau_2)}. \tag{16}$$

B. Markov approximation and single-atom density matrix equation

The effective single-atom time-evolution operator (13) leads in general to integro-differential equations of motion. We therefore restrict the discussion to situations that justify a Markov approximation, i.e. we assume that the characteristic decay time of field cumulants is short compared to the characteristic time of the atomic dynamics. This is the case, for example, if the medium is inhomogeneously broadened or under quasi-stationary conditions. We note that the Markov approximation used in the present paper rules out superradiance, since the superradiance time is shorter than the decay time of field correlations [11,12]. In order to describe fast *cooperative* processes, pair-correlations between different atoms need to be taken into account as a dynamical variable. This will be discussed in a future publication, where we derive an effective density matrix equation for atom pairs [20].

The Markov approximation amounts to assuming a δ-correlation of $D_{\mu\nu}$ and $C_{\mu\nu}$ in *physical* times.

$$D_{\mu\nu}^{AB}(\tau,\tau')=D_{\mu\nu}^{AB}(\tau)\,\delta(\tau-\tau'), \tag{17}$$
$$C_{\mu\nu}^{AB}(\tau,\tau')=C_{\mu\nu}^{AB}(\tau)\,\delta(\tau-\tau'), \tag{18}$$

with $A,B\in\{+,-\}$ explicitly denoting the contour branches. It is convenient to introduce dimensionless dipole operators $\sigma,\sigma^\dagger$, such that $p_\mu^+(t)=\wp_\mu\,\sigma_\mu(t)$ and $p_\mu^-(t)=\wp_\mu\,\sigma_\mu^\dagger(t)$ (and corresponding relations for the slowly-varying variables). With this we eventually arrive at

$$S_C^{\rm eff} T_C \exp\Bigg\{$$
$$\frac{i\wp_\mu}{\hbar}\int_{-\infty}^{\infty}d\tau\Big[\sigma_\mu(\tau_+)\mathcal{E}_{L\mu}^-(\vec r_0,\tau)-\sigma_\mu(\tau_-)\mathcal{E}_{L\mu}^-(\vec r_0,\tau)$$
$$+\sigma_\mu^\dagger(\tau_+)\mathcal{E}_{L\mu}^+(\vec r_0,\tau)-\sigma_\mu^\dagger(\tau_-)\mathcal{E}_{L\mu}^+(\vec r_0,\tau)\Big]$$
$$-\int_{-\infty}^{\infty}d\tau\,\frac{\Gamma_{\mu\nu}(\omega,\tau)}{2}\Big[\sigma_\mu(\tau_+)\sigma_\nu^\dagger(\tau_+)+$$
$$\sigma_\mu(\tau_-)\sigma_\nu^\dagger(\tau_-)-2\sigma_\mu(\tau_-)\sigma_\nu^\dagger(\tau_+)\Big] \tag{19}$$
$$-\int_{-\infty}^{\infty}d\tau\left(\frac{\Gamma_{\mu\nu}(\omega,\tau)}{2}+\frac{\gamma_{\mu\nu}(\omega,\tau)}{2}\right)\Big[\sigma_\nu^\dagger(\tau_+)\sigma_\mu(\tau_+)+$$
$$\sigma_\nu^\dagger(\tau_-)\sigma_\mu(\tau_-)-2\sigma_\nu^\dagger(\tau_-)\sigma_\mu(\tau_+)\Big]$$
$$+\frac{i}{\hbar}\int_{-\infty}^{\infty}d\tau\,H_{\mu\nu}(\omega,\tau)\Big[\sigma_\mu(\tau_+)\sigma_\nu^\dagger(\tau_+)-$$
$$\sigma_\mu(\tau_-)\sigma_\nu^\dagger(\tau_-)-\sigma_\nu^\dagger(\tau_+)\sigma_\mu(\tau_+)+\sigma_\nu^\dagger(\tau_-)\sigma_\mu(\tau_-)\Big]$$
$$+\frac{i}{\hbar}\int_{-\infty}^{\infty}d\tau\,h_{\mu\nu}(\omega,\tau)\Big[\sigma_\nu^\dagger(\tau_+)\sigma_\mu(\tau_+)-\sigma_\nu^\dagger(\tau_-)\sigma_\mu(\tau_-)\Big]\Bigg\}.$$

The lower indices $\pm$ at the time argument denote the branch on the Schwinger-Keldysh contour which is relevant for operator ordering under the action of T_C. The first term in (19) describes the interaction of the probe atom with the local field in RWA.

$$\Gamma_{\mu\nu}(\omega,t) = \frac{\wp_\mu\wp_\nu}{\hbar^2}\int_{-\infty}^{\infty} d\tau \,\langle\langle E_\mu^-(\vec{r}_0,t)E_\nu^+(\vec{r}_0,t+\tau)\rangle\rangle\, e^{i\omega\tau} = \frac{\wp_\mu\wp_\nu}{\hbar^2}\,\widetilde{D}_{\mu\nu}^{-+}(\vec{r}_0,\omega;t) \tag{20}$$

is a positive hermitian matrix, whose eigenvalues describe decay and pump rates induced by the incoherent photons inside the medium. Eq.(20) has a simple physical interpretation. The incoherent radiation inside the medium causes stimulated transitions from excited to ground states and vice versa. The corresponding rate is proportional to the spectral density of the radiation taken at the atomic transition frequency. Apart from some dimensional constants $\widetilde{D}_{\mu\nu}^{-+}$ is precisely the spectral energy density of the incoherent field at the position $\vec{r}_0$ and at the transition frequency ω of the probe atom.

$$\gamma_{\mu\nu}(\omega,t) = \frac{\wp_\mu\wp_\nu}{\hbar^2}\int_{-\infty}^{\infty} d\tau \Big\langle\big[E_\nu^+(\vec{r}_0,t+\tau),E_\mu^-(\vec{r}_0,t)\big]\Big\rangle\, e^{i\omega\tau} \tag{21}$$

is the spontaneous contribution to the "down rate" in the atomic medium. (Note that the commutator contains the field operators *interacting with the environment atoms.*) Since we are not interested here in the effects of the medium to the spontaneous decay, we replace $\gamma_{\mu\nu}$ by the free-space value $\gamma_{\mu\nu}^0$. We will show in Appendix A, that Eq.(21) indeed leads to the well-known Wigner-Weisskopf result for radiative decay in free space, if we replace E by the free field. Light-shifts induced by the incoherent component of the radiation field inside the medium are described by the hermitian matrix

$$H_{\mu\nu}(\omega,t) = \frac{i}{\hbar}\frac{\wp_\mu\wp_\nu}{2}\int_0^{\infty} d\tau\Big[\langle\langle E_\mu^-(\vec{r}_0,t)E_\nu^+(\vec{r}_0,t-\tau)\rangle\rangle\, e^{-i\omega\tau} - \langle\langle E_\mu^-(\vec{r}_0,t)E_\nu^+(\vec{r}_0,t+\tau)\rangle\rangle\, e^{+i\omega\tau}\Big]. \tag{22}$$

Eq.(22) can also be expressed in terms of D^{-+}:

$$H_{\mu\nu}(\omega,t) = \frac{\wp_\mu\wp_\nu}{2\pi\hbar}\,\mathrm{P}\!\int_{-\infty}^{\infty} d\omega' \;\frac{\widetilde{D}_{\mu\nu}^{-+}(\vec{r}_0,\omega';t)}{\omega-\omega'} = \frac{\hbar}{2\pi}\,\mathrm{P}\!\int_{-\infty}^{\infty} d\omega' \;\frac{\Gamma_{\mu\nu}(\omega',t)}{\omega-\omega'}, \tag{23}$$

where P denotes the principle part of the integral. In systems with inhomogeneous broadening the collective light-shifts are often negligible as they are usually small compared to the inhomogeneous width.

$$h_{\mu\nu}(\omega,t) = \frac{i}{\hbar}\frac{\wp_\mu\wp_\nu}{2}\int_0^{\infty} d\tau \tag{24}$$

$$\Big[\langle[E_\mu^-(\vec{r}_0,t),E_\nu^+(\vec{r}_0,t-\tau)]\rangle\, e^{-i\omega\tau} - \langle[E_\mu^-(\vec{r}_0,t),E_\nu^+(\vec{r}_0,t+\tau)]\rangle\, e^{i\omega\tau}\Big] \tag{25}$$

is the corresponding spontaneous contribution. Within the approximations made, $h_{\mu\nu}$ reflects the Lamb-shift of excited states altered by the presence of the medium. Here we are not interested in the Lamb shift and therefore consider it included in the free Hamiltonian H_0.

The effective time-evolution operator (19) directly leads to the following master equation for the single-atom density operator:

$$\begin{aligned}\dot{\rho} = &-\frac{i}{\hbar}\Big[H_0,\rho\Big] + i\frac{\wp_\mu}{\hbar}\Big[\sigma_\mu\mathcal{E}_{L\mu}^- + \sigma_\mu^\dagger\mathcal{E}_{L\mu}^+,\rho\Big]\\ &+\frac{i}{\hbar}H_{\mu\nu}\Big[\sigma_\mu\sigma_\nu^\dagger - \sigma_\nu^\dagger\sigma_\mu,\rho\Big]\\ &-\frac{\Gamma_{\mu\nu}}{2}\Big\{\sigma_\mu\sigma_\nu^\dagger\rho + \rho\sigma_\mu\sigma_\nu^\dagger - 2\sigma_\nu^\dagger\rho\sigma_\mu\Big\}\\ &-\left(\frac{\Gamma_{\mu\nu}}{2}+\frac{\gamma_{\mu\nu}^0}{2}\right)\Big\{\sigma_\nu^\dagger\sigma_\mu\rho + \rho\sigma_\nu^\dagger\sigma_\mu - 2\sigma_\mu\rho\sigma_\nu^\dagger\Big\}.\end{aligned} \tag{26}$$

This is the first main result of the present paper. We note that this equation is nonlinear and nonlocal, since the light-shift and decay matrices depend via the field correlations on the surrounding atoms. The equation does however have the Lindblad form [21] and thus preserves positivity and the total probability. In order to obtain a closed set of equations, we calculate in the following sections the yet unknown quantities $\mathcal{E}_L$, $\Gamma_{\mu\nu}$, and $H_{\mu\nu}$ in terms of single-atom density matrix elements.

III. THE AVERAGE LOCAL FIELD AND THE LORENTZ-LORENZ RELATION

We recognize from Eq.(26) that the probe atom is coupled to a classical (c-number) field of amplitude

$$\vec{\mathcal{E}}_L(\vec{r},t) = \vec{\mathcal{E}}(\vec{r},t) + \langle\vec{E}(\vec{r},t)\rangle. \tag{27}$$

The first term is the external coherent field (= field in the absence of the medium), and the second term is the mean coherent amplitude of the field scattered by all other atoms. Note, that the contribution of the probe atom itself is not included. On the other hand, the macroscopic field $\mathcal{E}_M$, which enters Maxwell's equations is the *total* field inside the medium (averaged over a spatial region large compared to the characteristic atomic distance, but smaller than λ^3). Thus the *local* field, given in (27) differs from the macroscopic Maxwell field essentially by the scattering contribution of the probe atom itself. In a continuum approximation we find

$$\mathcal{E}_{L\alpha}(\vec{r},t) = \mathcal{E}_{M\alpha}(\vec{r},t) - \frac{i}{\hbar}\varrho\int_{K_\epsilon} d^3\vec{r}\,'\int_{-\infty}^{\infty} dt'\, D_{0\,\alpha\beta}^{\mathrm{ret}}(\vec{r},t;\vec{r}\,',t')\,\langle p_{H\beta}(t')\rangle, \tag{28}$$

where $D_0^{\rm ret}$ is the free-field retarded propagator and $\langle p_H\rangle$ is the expectation value of the probe-dipole operator (in the Heisenberg picture). ϱ is the atomic density and K_ϵ denotes integration over a small sphere of radius ϵ. The retarded propagator of the electric field is given by [22]

$$D^{\rm ret}_{0\,\alpha\beta}(1,2) = \qquad (29)$$
$$\frac{i\hbar}{4\pi\epsilon_0 c}\Theta(\tau)\left[\delta_{\alpha\beta}\frac{\partial^2}{\partial\tau^2}-c^2\frac{\partial^2}{\partial x_2^\alpha\partial x_2^\beta}\right]\frac{\delta(r-c\tau)}{r},$$

where $\tau=t_1-t_2$, $r=|\vec r_1-\vec r_2|$ and Θ is the Heaviside step function.

When substituting $D_0^{\rm ret}$ from Eq.(29) into Eq.(28) we note that in the limit $\epsilon\to 0$ only the term which results from the second spatial derivative of $1/r$ survives. Using

$$\frac{\partial^2}{\partial x_2^\alpha\partial x_2^\beta}\;\frac{1}{r}=-\frac{4\pi}{3}\;\delta^{(3)}(\vec r_1-\vec r_2)\;\delta_{\alpha\beta}, \qquad (30)$$

we find

$$\vec{\mathcal{E}}_L(\vec r,t)=\vec{\mathcal{E}}_M(\vec r,t)+\frac{1}{3\epsilon_0}\varrho\Big\langle\vec p_H(t)\Big\rangle$$
$$=\vec{\mathcal{E}}_M(\vec r,t)+\frac{1}{3\epsilon_0}\vec{\mathcal{P}} \qquad (31)$$

which is identical to the classical Lorentz-Lorenz relation [1,2] when we identify $\vec{\mathcal{P}}=\varrho\langle\vec p_H\rangle$. It should be mentioned that the Lorentz-Lorenz relation holds for the mean amplitude of the field and not for the field operators itself as claimed in [23].

Making use of (31) we can define an effective semiclassical interaction operator

$$V_L=-\sum_j \vec p^{\,j}_\mu(t)\vec{\mathcal{E}}_{L\mu}(\vec r_j,t) \quad . \qquad (32)$$

IV. QUANTUM CORRECTIONS

We now discuss the light-shift and decay matrices in the generalized density-matrix equation (26) in more detail. Both depend on the field cumulants or Greensfunctions

$$D^{-+}_{\mu\nu}(\vec r,t;\vec r,t')=\langle\langle E^-_\mu(\vec r,t)E^+_\nu(\vec r,t')\rangle\rangle. \qquad (33)$$

(Note that the superscript "$-+$" indicates that the first time argument is on the lower and the second time argument on the upper branch of the Keldysh contour and has nothing to do with the frequency components of the field.) The aim of the present section is to calculate D^{-+} in terms of atomic variables. For this we apply non-equilibrium Greensfunction techniques [24].

1. Dyson-equation for $D(\check 1,\check 2)$

We define the exact and the (known) free Greensfunctions (GF) on the Keldysh contour as

$$D_{\mu\nu}(\check 1,\check 2)=\langle\langle T_C E^-_\mu(\vec r_1,\check t_1)E^+_\nu(\vec r_2,\check t_2)\rangle\rangle, \qquad (34)$$
$$D_{0\,\mu\nu}(\check 1,\check 2)=\langle\langle T_C E^-_{0\mu}(\vec r_1,\check t_1)E^+_{0\nu}(\vec r_2,\check t_2)\rangle\rangle, \qquad (35)$$

where E_0 denotes the free field, i.e. without coupling to the medium. The contour-Greensfunction $D(\check 1,\check 2)$ contains four real-time GFs: $D^{++}(1,2)$, $D^{-+}(1,2)$, $D^{+-}(1,2)$, and $D^{--}(1,2)$, where the superscripts "$\pm$" specify contour branches. The first and the last are the time- and anti-time ordered propagators and the retarded and advanced propagators are given by the combinations [24]

$$D^{\rm ret}(1,2)=D^{++}(1,2)-D^{+-}(1,2) \qquad (36)$$
$$=D^{-+}(1,2)-D^{--}(1,2),$$
$$D^{\rm adv}(1,2)=D^{++}(1,2)-D^{-+}(1,2) \qquad (37)$$
$$=D^{+-}(1,2)-D^{--}(1,2).$$

Within the RWA and in the absence of thermal photons, we have

$$D^{++}_{0\alpha\beta}(1,2)\approx D^{\rm adv}_{0\alpha\beta}(1,2), \qquad (38)$$
$$D^{-+}_{0\alpha\beta}(1,2)\approx 0, \qquad (39)$$
$$D^{+-}_{0\alpha\beta}(1,2)\approx D^{\rm adv}_{0\alpha\beta}(1,2)-D^{\rm ret}_{0\alpha\beta}(1,2), \qquad (40)$$
$$D^{--}_{0\alpha\beta}(1,2)\approx -D^{\rm ret}_{0\alpha\beta}(1,2). \qquad (41)$$

A formal solution to the atom-field interaction can be given in terms of a Dyson-integral equation [24], by introducing a formal polarization function $\Pi_{\alpha\beta}(\check 1,\check 2)$

$$D_{\mu\nu}(\check 1,\check 2)\;=\;D_{0\,\mu\nu}(\check 1,\check 2)-$$
$$\iint_C d\check 1'\,d\check 2'\,D_{0\,\mu\alpha}(\check 1,\check 1')\,\Pi_{\alpha\beta}(\check 1',\check 2')\,D_{\beta\nu}(\check 2',\check 2). \qquad (42)$$

Here $\int_C d\check 1$ denotes integration over the Schwinger-Keldysh contour as well as spatial integration over the medium. The Dyson-equation (42) represents nothing else than a formal summation of the perturbation series where the polarization function is determined by the medium response. We now have to find a good approximation for $\Pi(1,2)$.

2. self-consistent Hartree approximation

One easily verifies that in lowest order in the atom-field coupling, the polarization function is given by a correlation function of dipole operators of non-interacting atoms

$$\Pi^{(0)}_{\alpha\beta}(\check 1,\check 2)\;= \qquad (43)$$
$$\frac{\wp_\alpha\wp_\beta}{\hbar^2}\sum_j\Big\langle\Big\langle T_C\sigma^\dagger_{j\alpha}(\check t_1)\sigma_{j\beta}(\check t_2)\Big\rangle\Big\rangle_{\rm free}\delta(\vec r_1-\vec r_j)\,\delta(\vec r_2-\vec r_j).$$

This corresponds to a Hartree approximation in many-body theory. This approximation is physically justified, when the nonlinear light-shift and decay terms do not affect the atomic dynamics, that is if the probability that a specific atom reabsorbs or scatters a spontaneous photon is small. Such a situation is realized, for example, in the classical case of radiation trapping where a small number of photons (much smaller than necessary to saturate the medium) is trapped in a dense absorbing medium [13]. We are here also interested, however, in situations, where incoherent photons significantly alter the atomic dynamics. A consistent approximation, which accounts also for these cases is the self-consistent Hartree approximation, where the cumulants of *free* dipole operators in (43) are replaced by cumulants of *interacting* dipole operators.

$$\Pi_{\alpha\beta}(\check{1},\check{2}) = \frac{\wp_\alpha\wp_\beta}{\hbar^2}\sum_j \langle\langle T_C\sigma^\dagger_{j\alpha}(\check{t}_1)\sigma_{j\beta}(\check{t}_2)\rangle\rangle\,\delta(\vec{r}_1-\vec{r}_j)\,\delta(\vec{r}_2-\vec{r}_j). \quad (44)$$

As shown in Appendix B the Dyson-equation for the contour GF can be rewritten in the RWA in terms of the real-time GFs as follows:

$$D^{-+}_{\alpha\beta}(1,2) = -\iint d3\,d4\,D^{\rm ret}_{\alpha\mu}(1,3)\,\Pi^{\rm s}_{\mu\nu}(3,4)\,D^{\rm adv}_{\nu\beta}(4,2), \quad (45)$$

where $D^{\rm ret}_{\mu\nu}(1,2)\,\big(=D^{\rm adv}_{\nu\mu}(2,1)\big)$ obeys the Dyson equation

$$D^{\rm ret}_{\alpha\beta}(1,2) = D^{\rm ret}_{0\,\alpha\beta}(1,2) - \iint d3\,d4\,D^{\rm ret}_{0\,\alpha\mu}(1,3)\,\Pi^{\rm ret}_{\mu\nu}(3,4)\,D^{\rm ret}_{\nu\beta}(4,2). \quad (46)$$

Here the time integration goes from $-\infty$ to ∞ and we have introduced the atomic source correlation

$$\Pi^{\rm s}_{\mu\nu}(\vec{r}_1,t_1;\vec{r}_2,t_2) = \frac{\wp_\mu\wp_\nu}{\hbar^2}\sum_j\big\langle\big\langle\sigma^\dagger_{j\mu}(t_1)\sigma_{j\nu}(t_2)\big\rangle\big\rangle\,\delta(\vec{r}_1-\vec{r}_j)\,\delta(\vec{r}_2-\vec{r}_j) \quad (47)$$

as well as the atomic response function

$$\Pi^{\rm ret}_{\mu\nu}(\vec{r}_1,t_1;\vec{r}_2,t_2) = \frac{\wp_\mu\wp_\nu}{\hbar^2}\Theta(t_1-t_2)\sum_j\big\langle\big[\sigma^\dagger_{j\mu}(t_1),\sigma_{j\nu}(t_2)\big]\big\rangle\,\delta(\vec{r}_1-\vec{r}_j)\,\delta(\vec{r}_2-\vec{r}_j). \quad (48)$$

The names reflect the physical meaning of the terms. The Fourier-transform of $\Pi^{\rm s}$ is proportional to the spontaneous emission spectrum of the atoms and that of $\Pi^{\rm ret}$ gives the susceptibility of the medium.

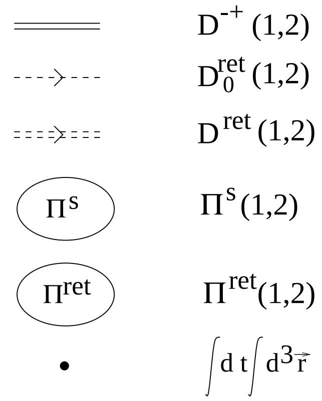

FIG. 2. Feynman diagrams, definitions

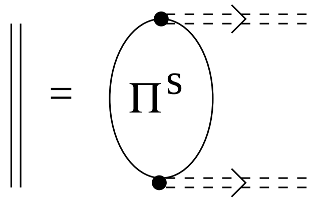

FIG. 3. Graphical representation of Eq.(45). The incoherent intensity at the position of the probe atom is the sum of all spontaneous contribution propagated through the medium.

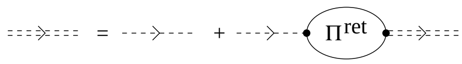

FIG. 4. Graphical representation of Dyson equation (46) for retarded GF inside the medium. Iteration generates all-order scattering contributions.

Eqs.(45) and (46) can be given an instructive graphical representation shown in Figs. 2-4. Eq.(45) (illustrated in Fig. 3) says that the incoherent radiation intensity is obtained by summing the spontaneous-emission contributions from all atoms propagated through the medium. The iteration of the Dyson equation (46) (shown in Fig. 4) describes multiple scattering of spontaneous photons by atoms during the propagation from a source atom to the probe atom.

3. Explicit expressions for the collective decay rate and light-shift

We now approximately solve the Dyson-equation (46) for the retarded propagator in the medium. We first introduce a continuum approximation.

$$\Pi^{\rm ret}_{\mu\nu}(\vec{r}_1,t_1;\vec{r}_2t_2) = \int d^3\vec{r}\;P^{\rm ret}_{\mu\nu}(\vec{r},t_1,t_2)\,\delta(\vec{r}_1-\vec{r})\,\delta(\vec{r}_2-\vec{r}), \quad (49)$$

$$P^{\rm ret}_{\mu\nu}(\vec{r}_j,t_1,t_2) = \frac{\wp_\mu\wp_\nu}{\hbar^2}N\Theta(t_1-t_2)\,\overline{\langle[\sigma^\dagger_{j\mu}(t_1),\sigma_{j\nu}(t_2)]\rangle}\,, \quad (50)$$

where N is the atom density and the overline denotes averaging over some inhomogeneous distribution. Similarly

$$\Pi^{\rm s}_{\mu\nu}(\vec r_1,t_1;\vec r_2,t_2) = \int d^3\vec r\ P^{\rm s}_{\mu\nu}(\vec r,t_1,t_2)\,\delta(\vec r_1-\vec r)\,\delta(\vec r_2-\vec r), \tag{51}$$

$$P^{\rm s}_{\mu\nu}(\vec r_j,t_1,t_2) = \frac{\wp_\mu\wp_\nu}{\hbar^2}N\ \overline{\langle\langle\sigma^\dagger_{j\mu}(t_1)\sigma_{j\nu}(t_2)\rangle\rangle} \tag{52}$$

Thus Eq.(46) reads

$$D^{\rm ret}_{\alpha\beta}(\vec r_1,t_1;\vec r_2,t_2) = D^{\rm ret}_{0\,\alpha\beta}(\vec r_1,t_1;\vec r_2,t_2)- \int_{-\infty}^{\infty}dt'_1\int_{-\infty}^{\infty}dt'_2\int_V d^3\vec r\,'_1\ D^{\rm ret}_{0\,\alpha\mu}(\vec r_1,t_1;\vec r\,'_1,t'_1)\ P^{\rm ret}_{\mu\nu}(\vec r\,'_1;t'_1,t'_2)\ D^{\rm ret}_{\nu\beta}(\vec r\,'_1,t'_2;\vec r_2,t_2). \tag{53}$$

To solve this integral equation, we now make the following approximations. We first extend the spatial integration to infinity, which basically means that we are solving for the retarded propagator in an infinitely extended medium. Secondly, we replace $\vec r\,'_1$ in the atomic response function by $\vec r_2$, i.e. we evaluate the response at the position of the source. We furthermore consider quasi-stationary conditions, i.e. assume that $P^{\rm ret}(t'_1,t'_2)$ depends only on the time difference $\tau = t'_1 - t'_2$. We only keep an overall slow (parametric) time dependence. This means that we consider propagation times short compared to the characteristic time of the atomic evolution, which is consistent with the earlier Markov approximation. With these simplifications we can turn the integral equation (53) into an algebraic one by Fourier-transformation with respect to $\vec x\equiv\vec r_1-\vec r_2$ and $\tau\equiv t_1-t_2$. At this point a word of caution is needed: As will be discussed in Appendix C, the retarded GF in an *amplifying* medium is not Fourier-transformable, since it grows exponentially with $r=|\vec x|$. We therefore should view the transformations as finite-time and finite-space Fourier-transforms, and hence the algebraic equation as an approximation.

Using the definition

$$\widetilde{\widetilde{\rm F}}(\vec q,\omega) = \int_{V_\infty} d^3\vec x\int_{-\infty}^{\infty}d\tau\,{\rm F}(\vec x,\tau)\,e^{-i\omega\tau}\,e^{i\vec q\cdot\vec x}, \tag{54}$$

the solution of Eq.(53) reads

$$\widetilde{\widetilde{\mathbf D}}^{\rm ret}(\vec q,\omega;t) = \Big[\mathbf 1 + \widetilde{\widetilde{\mathbf D}}_0^{\rm ret}(\vec q,\omega)\cdot\widetilde{\mathbf P}^{\rm ret}(\omega;t)\Big]^{-1}\cdot\widetilde{\widetilde{\mathbf D}}_0^{\rm ret}(\vec q,\omega). \tag{55}$$

Here $\mathbf D^{\rm ret}$ and $\mathbf\Pi^{\rm ret}$ denote 3×3 matrices in coordinate space and $\mathbf 1$ is the unity matrix.

For simplicity we now disregard polarization, i.e. we replace the 3×3 matrices by simple functions. We note however, that a generalization is straight forward. As shown in detail in Appendix C, we eventually arrive at

$$\widetilde D^{\rm ret}(\vec x,\omega;t) = -\frac{i\hbar\omega^2}{6\pi\epsilon_0c^2}\frac{e^{q''_0r}}{r}\,e^{-iq'_0r} \tag{56}$$

where λ is the wavelength of the transition under consideration in the rest frame, and $r=|\vec x|=|\vec r_1-\vec r_2|$.

$$q_0 = q'_0(\vec r,\omega,t)+iq''_0(\vec r,\omega,t) = \frac{\omega}{c}\left[1+\frac{i\hbar}{3\epsilon_0}\widetilde P^{\rm ret}(\vec r,\omega;t)\right]. \tag{57}$$

q''_0 is the inverse absorption/amplification length in the medium and q'_0 characterizes the corresponding phase shift. We here have assumed that $\big|{\rm Im}[\widetilde P^{\rm ret}]\big|\,\hbar/3\epsilon_0<1$.

With Eq.(56) we can now express $\widetilde D^{-+}(\vec r_0,\omega;t)$ in terms of atomic variables

$$\widetilde D^{-+}(\vec r_0,\omega,t) = \frac{\hbar^2\omega^4}{(6\pi)^2\epsilon_0^2c^4}\int_V d^3\vec r\ \frac{e^{2q''_0(\vec r,\omega;t)r}}{r^2}\ \widetilde P^{\rm s}(\vec r,\omega;t). \tag{58}$$

Here $r=|\vec r-\vec r_0|$ is the distance between source and probe atom. With Eq.(58) we finally find for the collective decay rate and light-shift

$$\Gamma(\omega,t) = \frac{\wp^2\omega^4}{(6\pi)^2\epsilon_0^2c^4}\int_V d^3\vec r\,\frac{e^{2q''_0(\vec r,\omega;t)r}}{r^2}\ \widetilde P^{\rm s}(\vec r,\omega;t), \tag{59}$$

$$H(\omega,t) = \frac{\hbar\wp^2\omega^4}{(6\pi)^2\epsilon_0^2c^4}\int_V d^3\vec r\ {\rm P}\!\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\ \frac{e^{2q''_0(\vec r,\omega';t)r}}{r^2}\ \frac{\widetilde P^{\rm s}(\vec r,\omega;t)}{\omega-\omega'}. \tag{60}$$

Eqs.(59) and (60) are the second major result of the present paper. In applying these results to a specific problem, we still have to calculate the source-correlation $P^{\rm s}$ in terms of density matrix elements. This then yields a closed nonlinear and nonlocal density matrix equation. We will illustrate this for some examples in the following section.

V. EXAMPLES

A. Inhomogeneously broadened two-level system

We here consider an inhomogeneously broadened dense ensemble of randomly polarized two-level atoms in a cylindrical geometry as shown in Fig. 5. For this system the time-evolution of the dipole operator $\sigma=|b\rangle\langle a|$ is determined by the simple Heisenberg-Langevin equation

$$\dot\sigma_j = -(i\omega^j_{ab}+\Gamma_{ab})\sigma_j + \text{noise}, \tag{61}$$

where the noise term denotes a white noise source, which is however of no interest here. $\omega^j_{ab}=\omega^0_{ab}+\Delta_j$ is the atomic transition frequency in the laboratory frame. We here take into account Doppler-broadening which leads to a shift Δ_j of the lab-frame transition frequency from the rest-frame frequency ω_{ab}^0. The collective light-shift is small compared to the average Doppler-shift and therefore neglected. The coherence decay rate Γ_{ab} consists of two contributions, one resulting from free-space spontaneous decay γ and the other from the collective decay Γ, $\Gamma_{ab} = \Gamma + \gamma/2$. Eq.(61) can easily be solved by Laplace-transformation ($\tilde{x}(s,t) := \int_0^\infty d\tau\, e^{-s\tau}\, x(t+\tau)$), which yields

$$\langle\langle\tilde{\sigma}_j^\dagger(s;t)\sigma_j(t)\rangle\rangle = \frac{\rho_{aa}^j(t)}{s - i\omega_{ab}^j + \Gamma_{ab}}, \tag{62}$$

$$\langle\langle\tilde{\sigma}_j(s;t)\sigma_j^\dagger(t)\rangle\rangle = \frac{\rho_{bb}^j(t)}{s + i\omega_{ab}^j + \Gamma_{ab}}. \tag{63}$$

From this we immediately obtain

$$\widetilde{P}^{\mathrm{ret}}(\vec{r}_j,\omega,t) = \frac{\wp^2}{\hbar^2} N\, \overline{\frac{\rho_{aa}^j(t) - \rho_{bb}^j(t)}{\Gamma_{ab} + i(\omega - \omega_{ab}^j)}}, \tag{64}$$

$$\widetilde{P}^{s}(\vec{r}_j,\omega,t) = \frac{2\wp^2}{\hbar^2} N\, \overline{\frac{\rho_{aa}^j(t)\Gamma_{ab}}{(\Gamma_{ab})^2 + (\omega - \omega_{ab}^j)^2}}, \tag{65}$$

where $\wp$ is the dipole moment of the transition and the overbar denotes averaging over the velocity distribution of the atoms, which is given by the Gaussian distribution

$$W(\Delta_j) = \frac{1}{\sqrt{2\pi}\Delta_D} \exp\left\{-\frac{\Delta_j^2}{2\Delta_D^2}\right\}. \tag{66}$$

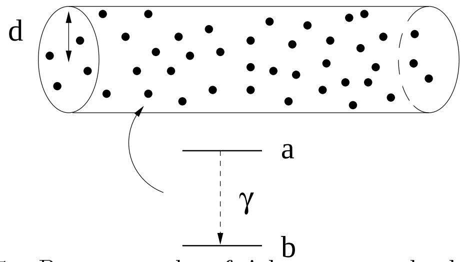

FIG. 5. Dense sample of inhomogeneously broadened two-level atoms in cylindrical geometry.

Since the lab-frame atomic transition frequency depends on the velocity, the collective decay rate, which is proportional to the incoherent radiation spectrum at this frequency, will be velocity dependent as well. Thus we have in general a set of nonlinear coupled equations corresponding to different velocity classes. If there are fast velocity-changing collision, the population dynamics of all velocity classes will however be approximately the same. In this case we may set

$$\rho_{\mu\mu}^j(t) = \overline{\rho_{\mu\mu}^j(t)} =: \rho_{\mu\mu}(\vec{r},t), \tag{67}$$

where $\mu \in \{a,b\}$ and $\vec{r}$ denotes the position of the atoms considered. Note, however, that this approximation does not hold if the inhomogeneous broadening mechanism is not due to Doppler as for example in solids. In that case one has to consider the full set of equations. Using Eq.(67), we find in the limit of large Doppler-broadening $\Delta_D \gg \Gamma_{ab}$

$$\widetilde{P}^{\mathrm{ret}}(\vec{r},\omega,t) = \frac{\wp^2 N}{\hbar^2}\sqrt{2\pi}\,\frac{\rho_{aa}(\vec{r},t) - \rho_{bb}(\vec{r},t)}{\Delta_D} e^{-\Delta^2/2\Delta_D^2}\left[1 - i\sqrt{\frac{2}{\pi}}\int_0^{\Delta/\Delta_D} dy\, e^{-y^2/2}\right], \tag{68}$$

where $\Delta = \omega - \omega_{ab}^0$ is the detuning from the atomic resonance at rest. For the collective decay rate only the real part of $\widetilde{P}^{\mathrm{ret}}$ is important which enters the absorption coefficient according to Eq.(57)

$$q_0''(\vec{r},\omega,t) = \frac{\wp^2 N}{3\hbar\epsilon_0}\frac{\omega}{c}\sqrt{2\pi}\,\frac{\rho_{aa}(\vec{r},t) - \rho_{bb}(\vec{r},t)}{\Delta_D}\, e^{-\Delta^2/2\Delta_D^2}. \tag{69}$$

Similarly we have for the source term in the strong Doppler-limit

$$\widetilde{P}^{s}(\vec{r},\omega,t) = \frac{2\wp^2 N}{\hbar^2}\sqrt{2\pi}\,\frac{\rho_{aa}(\vec{r},t)}{\Delta_D}\, e^{-\Delta^2/2\Delta_D^2}. \tag{70}$$

Combining (69) and (70) and applying the relation between the free-space radiative decay rate γ and the dipole moment $\wp$: $\wp^2 = 3\pi\hbar\epsilon_0 c^3\gamma/\omega^3$ [25] (cf. also Appendix A) yields the collective decay rate (59) for a probe atom with (lab-frame) transition frequency ω at position $\vec{r}_0$

$$\Gamma(\omega,t) = \gamma\int_V d^3\vec{r}\; 2q_0''(\vec{r},\omega,t)\,\frac{e^{2q_0''(\vec{r},\omega,t)r}}{4\pi r^2}\,\frac{\rho_{aa}(\vec{r},t)}{\rho_{aa}(\vec{r},t) - \rho_{bb}(\vec{r},t)}, \tag{71}$$

with $r = |\vec{r} - \vec{r}_0|$. To obtain the effective decay/pump rate we have to averaged over the velocity distribution

$$\Gamma(t) = \overline{\Gamma(\omega,t)} = \int_{-\infty}^{\infty} d\omega\, \frac{1}{\sqrt{2\pi}\Delta_D} e^{-(\omega-\omega_{ab}^0)^2/2\Delta_D^2}\,\Gamma(\omega,t). \tag{72}$$

We now discuss two limiting cases. In the first case we assume a small excitation in the medium. This corresponds to the classical situation of radiation trapping in an inhomogeneously broadened two-level medium. We will show that in this case Eq.(72) leads to the integral equation of Holstein [13]. In the second case we will disregard the spatial dependence but keep the nonlinearities, and consider the temporal evolution from an initially excited ensemble.

1. linear limit and Holstein equations of radiation trapping

For small excitation, the retarded light propagation can be regarded as propagation in a medium with all population in the lower state, i.e. $\rho_{bb}=1$ and $\rho_{aa}=0$. Thus

$$q_0''(\vec{r},\omega,t)=q_0''(\omega,t)=-\frac{\wp^2 N}{3\hbar\epsilon_0}\frac{\omega}{c}\frac{\sqrt{2\pi}}{\Delta_D}\,e^{-\Delta^2/2\Delta_D^2} \tag{73}$$

and we can approximate the denominator in (71) by -1. This results in

$$\Gamma(t)\approx\gamma\int_V d^3\vec{r}\;G(\vec{r}_0,\vec{r})\,\rho_{aa}(\vec{r},t) \tag{74}$$

where

$$\begin{aligned} G(\vec{r}_0,\vec{r}) &= \qquad\qquad (75)\\ -\,2q_0''(\omega,t)\,\frac{e^{2q_0''(\omega,t)r}}{4\pi r^2} &= \\ -\int_{-\infty}^{\infty}d\omega\,\frac{1}{\sqrt{2\pi}\Delta_D}e^{-(\omega-\omega_{ab}^0)^2/2\Delta_D^2}\,2q_0''(\omega,t)\,\frac{e^{2q_0''(\omega,t)r}}{4\pi r^2} &= \\ \frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}dx\,e^{-x^2}\left(-\frac{1}{4\pi r^2}\right)\frac{\partial}{\partial r}\exp\Big[-K_0 e^{-x^2}r\Big]. \end{aligned}$$

Here $K_0=N\lambda^2 g$ and $g=\gamma/\sqrt{2\pi}\Delta_D$ characterizes the ratio of the homogeneous to the inhomogeneous width.

The dynamical evolution of the ensemble is described by the Bloch equation

$$\dot\rho_{aa}(\vec{r}_0,t)=\Gamma(t)-\Big[\gamma+2\Gamma(t)\Big]\rho_{aa}(\vec{r}_0,t). \tag{76}$$

In the small-excitation limit, the term $\Gamma\rho_{aa}$ is of second order and can be neglected. We thus arrive at the *linear* integral equation for the atomic excitation

$$\begin{aligned} \dot\rho_{aa}(\vec{r}_0,t) &= \qquad\qquad (77)\\ -\gamma\,\rho_{aa}(\vec{r}_0,t) &+ \gamma\int_V d^3\vec{r}\;G(\vec{r}_0,\vec{r})\,\rho_{aa}(\vec{r},t). \qquad (78) \end{aligned}$$

Eq.(77) is the integro-differential equation for radiation trapping derived by Holstein in [13] for the special case of Doppler-broadened two-level atoms. Thus in the linear limit we have rederived the theory of radiation trapping of [13].

2. dynamics of initially inverted two-level system in small-sample approximation

Let us now discuss a nonlinear problem, but in a small volume, such that the space dependence can be disregarded. In this case we can carry out the volume integral placing the probe atom on the axis of the long cylindrical sample (see Fig.5). We find for the decay rate for a probe atom with transition frequency ω

$$\frac{\Gamma(\omega,t)}{\gamma}=\frac{\rho_{aa}(t)}{\rho_{bb}(t)-\rho_{aa}(t)}\left[1-\exp\left(-K(t)e^{-\Delta^2/2\Delta_D^2}\right)\right], \tag{79}$$

where $\Delta=\omega-\omega_{ab}^0$, and

$$K(t)=K_0 d\,[\rho_{bb}(t)-\rho_{aa}(t)]. \tag{80}$$

Averaging over the inhomogeneous velocity distribution of the atoms yields

$$\begin{aligned} \frac{\Gamma(t)}{\gamma} &= \frac{\rho_{aa}(t)}{\rho_{bb}(t)-\rho_{aa}(t)} \\ &\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}dy\,e^{-y^2}\left[1-\exp\left(-K(t)e^{-y^2}\right)\right]. \qquad (81) \end{aligned}$$

Note that $\Gamma(t)/\gamma$ remains finite at $\rho_{aa}=1/2$, since the diverging denominator is multiplied by a vanishing integral expression.

The time evolution of the excited-state population from an initially completely inverted system is shown in Fig. 6 for different values of the density $\eta\equiv N\lambda^2 d=100$ (solid line) and $\eta=500$ (dashed line) and $g=0.01$. The dotted line corresponds to the free-space decay. One recognizes a non-exponential behavior, with an accelerated decay in the initial phase corresponding to amplified spontaneous emission and a substantial slow-down of decay in the long-time limit.

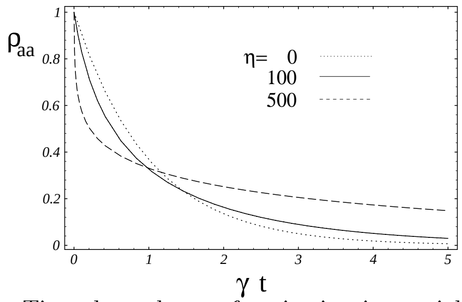

FIG. 6. Time dependence of excitation in spatially homogeneous, dense two-level medium. Time is in units of the inverse free-space decay rate. $\eta\equiv N\lambda^2 d=0$ (dotted), 100 (line), 500 (dashed). $g=\gamma/\sqrt{2\pi}\Delta_D=0.01$.

The effective rate of decay of the excitation $\Gamma_{\rm eff}=-\dot\rho_{aa}/\rho_{aa}$ is shown in Fig. 7. One can see that for $\eta=500$ the initial decay rate is already of the order of the inhomogeneous Doppler-width ($\log\Delta_D/\gamma\approx 1.6$) and the Markov-approximation of slow atomic evolution becomes invalid. For higher atomic densities the system would show superradiant decay in the initial phase which cannot be described by the single-atom density matrix equation. As noted before, modeling of the cooperative decay requires a two-atom density matrix description, which will be discussed elsewhere [20].

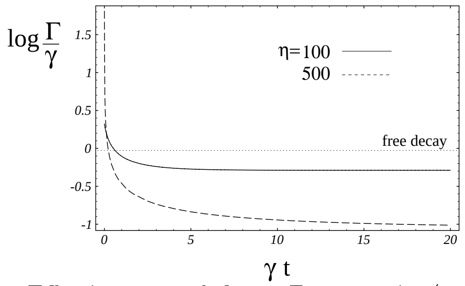

FIG. 7. Effective rate of decay $\Gamma_{\rm eff} = -\dot\rho_{aa}/\rho_{aa}$ for examples of Fig.6. Dotted line corresponds to free-space decay. Amplified spontaneous emission in the initial phase and slow-down of decay in final phase are apparent.

One also verifies from Fig. 7 for the case $\eta = 100$, that the decay becomes exponential again in the long-time limit. The asymptotic escape rate is given by

$$\gamma_{\rm esc} = \frac{\gamma}{K_0(\pi \ln K_0)^{1/2}}, \tag{82}$$

which can be orders of magnitude smaller than γ. This result agrees with Eq.(1.1) of [13]b up to a numerical factor of the order of unity, which is due to the fact that we here have disregarded a possible spatial inhomogeneity.

It is also instructive to consider the time-dependent spectrum of incoherent radiation or equivalently $\Gamma(\omega, t)$. This is done in Fig. 8 for $\eta = 500$. Shown is the spectral distribution at different times normalized to the averaged rate $\Gamma(t)$.

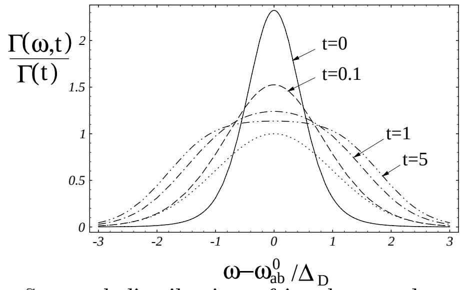

FIG. 8. Spectral distribution of incoherent decay as function of time. $\eta = 500$, $g = 0.01$. Dotted line shows Doppler-distribution of atomic transition frequencies (not normalized). Time is in units of γ^{-1}.

The dotted line shows the (not normalized) inhomogeneous distribution of atomic frequencies according to Eq.(66). One recognizes that the incoherent spectrum broadens with the decay of excitation. In the initial phase of amplified spontaneous emission ($\gamma t = 0 \ldots 1$) one can see that the radiation spectrum is narrower than the inhomogeneous atomic spectrum. This gives the first indication of spectral condensation, a well-known phenomenon in amplifying media and lasers.

B. Effects of radiative atom-atom interactions on intrinsic optical bistability

One of the most interesting dynamical effects in dense media due to the Lorentz-Lorenz nonlinearity is the possibility of intrinsic optical bistability predicted in [7]. If a radiatively broadened two-level system is resonantly driven by a coherent field of Rabi-frequency Ω it shows mirrorless, i.e. intrinsic bistability, if the atomic density exceeds some critical value. The bistability results from an effective feedback introduced by the Lorentz-Lorenz correction.

We here consider a dense ensemble of resonantly driven two-level systems as shown in Fig. 9. For simplicity of the present discussion we assume that the driving field is homogeneous.

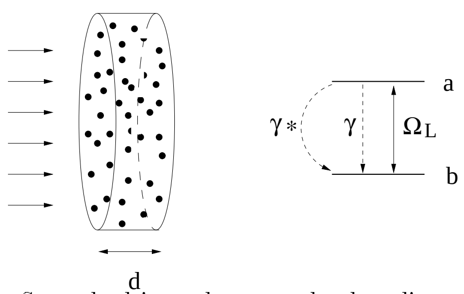

FIG. 9. Strongly driven, dense two-level medium. γ and γ^* describe radiative and non-radiative decays respectively, and Ω_L denotes Rabi-frequency of local field.

For most practical realizations this assumption is not valid. We are here however interested in principle questions and will therefore ignore drive-field depletion. The assumption of a homogeneous driving field implies a homogeneous behavior of the atomic system and we can disregard the spatial dependence in the collective decay and light-shift terms. The density matrix equations for the system under consideration read in a rotating frame

$$\dot\rho_{aa} = -\Gamma_a\rho_{aa} + \Gamma\rho_{bb} - i(\Omega\rho_{ab} - \Omega\rho_{ab}^*), \tag{83}$$

$$\dot\rho_{ab} = -\Gamma_{ab}\rho_{ab} - i(\Omega + C\gamma\rho_{ab})(\rho_{aa} - \rho_{bb}), \tag{84}$$

where we have assumed a real Ω. $\Gamma_a = \gamma + \gamma^* + \Gamma$ is the total population decay rate out of the excited state, with γ and γ^* being the free-space radiative and non-radiative decay rates, and Γ the collective decay rate. $\Gamma_{ab} = \Gamma + (\gamma + \gamma^*)/2$. There is no collective light-shift contribution here due to symmetry reasons. One recognizes a term proportional to the atomic polarization ρ_{ab} that adds to the Rabi-frequency Ω. This term is due to the Lorentz-Lorenz correction (31) and has the character of a feedback (atomic polarization generates a field contribution $C\gamma\rho_{ab}$ which couples back to the atom). $C = N\lambda^3/4\pi^2$ is the cooperativity parameter, that essentially determines the number of atoms in a volume λ^3.

The stationary solution of Eqs.(83) and (84) for the excited state population for $\Gamma = \gamma^* = 0$ is shown in Fig. 10

for different cooperativities. One recognizes bistability for $C \geq 3$.

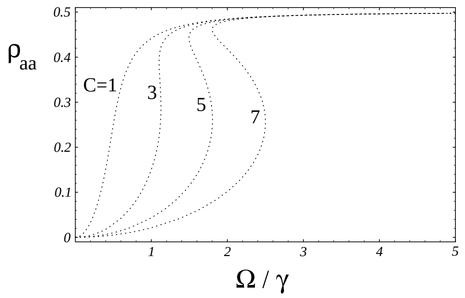

FIG. 10. Stationary excited state population as function of driving-field Rabi-frequency Ω for different cooperativity parameters. Here $\Gamma = \gamma^* = 0$.

We now analyze the effect of incoherent photons inside the medium. To self-consistently determine the collective decay rate, we have to calculate correlations of the dipole operators $\sigma = |b\rangle\langle a|$ and $\sigma^\dagger$ in terms of density matrix elements. More precisely we need the second order cumulants, i.e. correlations of operators minus their mean values (which are nonzero in the present case). Thus we start with the Heisenberg-Langevin equations for $\delta\sigma \equiv \sigma - \langle\sigma\rangle$ and $\delta\sigma_{aa} = \sigma_{aa} - \langle\sigma_{aa}\rangle$, where $\sigma_{aa} = |a\rangle\langle a|$:

$$\delta\dot{\sigma} = -\Gamma_{ab}\delta\sigma - 2i\Omega_L\delta\sigma_{aa} + \text{noise} \tag{85}$$

$$\delta\dot{\sigma}_{aa} = -2\Gamma_{ab}\delta\sigma_{aa} - i\Big(\Omega_L^*\delta\sigma - \Omega_L\delta\sigma^\dagger\Big) + \text{noise}, \tag{86}$$

where $\Omega_L = \Omega + C\gamma\rho_{ab}$. The relevant correlations can be obtained from these equations by Laplace-transformation. This yields at resonance ($\omega = \omega_{ab}$)

$$\widetilde{P}^{\rm ret}(\omega_{ab}) = \frac{\wp^2 N}{\hbar^2}\frac{\Gamma_{ab}\big(\rho_{aa} - \rho_{bb}\big)}{\Gamma_{ab}^2 + 2|\Omega_L|^2}, \tag{87}$$

$$\widetilde{P}^{\,\rm s}(\omega_{ab}) = \frac{2\wp^2 N}{\hbar^2}\frac{\Gamma_{ab}^2\Big(\rho_{aa} - |\rho_{ab}|^2\Big) + 2|\Omega_L|^2\rho_{aa}\rho_{bb}}{\Gamma_{ab}\Big(\Gamma_{ab}^2 + 2|\Omega_L|^2\Big)}. \tag{88}$$

Note that we have omitted space and time arguments, since we are interested in the stationary properties of the system in a homogeneous sample. From (87) we immediately find the absorption coefficient

$$q_0'' = \frac{\wp^2 N}{3\hbar\epsilon_0}\frac{\omega_{ab}}{c}\frac{\Gamma_{ab}}{\Gamma_{ab}^2 + 2|\Omega_L|^2}\big(\rho_{aa} - \rho_{bb}\big) \tag{89}$$

We now consider a thin plate of thickness d as shown in Fig. 9 and assume that the beam diameter of the driving field is large compared to d. Carrying out the spatial integrations in Eq.(59) using (89) and (88) we find the following relation for the collective decay rate

$$\frac{\Gamma}{\gamma} = \tag{90}$$

$$\frac{1}{\rho_{bb} - \rho_{aa}}\left(\rho_{aa} - |\rho_{ab}|^2 + \frac{2|\Omega_L|^2}{\Gamma_{ab}^2}\rho_{aa}\rho_{bb}\right)\left(1 - e^{2q_0'' d}\right).$$

This equation for Γ is not yet explicit since Γ enters the right hand side of the equation in an essentially nonlinear way. Using the stationary solutions of the density matrix equations (83) and (84) with Γ as independent variable, one can (with some additional approximations) solve Eqs.(90). This yields

$$\frac{\Gamma}{\gamma} \approx \frac{\rho_{aa}}{1 - 2\rho_{aa}}\Big(1 - e^{-K}\Big), \tag{91}$$

with

$$K = Cr\,\frac{1 + \gamma^*/\gamma(1 - 2\rho_{aa})}{2\frac{\Omega^2}{\gamma^2} + 2C^2(1 - 2\rho_{aa})^2 + \frac{(1+\gamma^*/\gamma(1-2\rho_{aa}))^2}{4(1-2\rho_{aa})^2}}. \tag{92}$$

C is the cooperativity parameter, and $r = \pi d/\lambda$.

In Fig. 11 we show the stationary solutions for the excited state population as function of the driving-field Rabi-frequency Ω for different cooperativity parameters and for purely radiative decay, i.e. $\gamma^* = 0$. The dotted curves correspond to the solutions without radiative atom-atom interactions. As can be seen, bistability persists, but cannot be resolved for physically reasonable values of r ($r = 100$ in Fig. 11b). Radiation trapping prevents the energy to escape from the sample and already very small external pumping is sufficient to keep the atoms in a highly excited state.

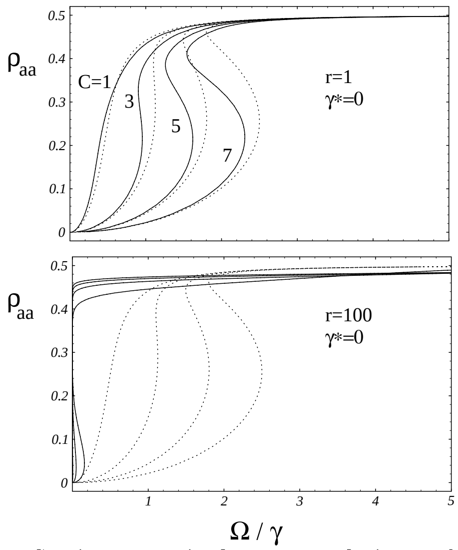

FIG. 11. Stationary excited state population as function of driving-field Rabi-frequency Ω for radiative decay ($\gamma^* = 0$) and with radiative atom-atom interactions. $r = \pi d/\lambda = 1$ (upper picture) and $r = 100$ (lower picture). Dotted curves show behavior without radiative atom-atom interactions (see Fig. 10.)

The situation is different, if there is also non-radiative decay, as shown in Fig. 12 (here $\gamma^* = \gamma$). In comparison to the radiatively-broadened case of Fig. 11, the bistability curves are only moderately altered even for large samples with $r = 1000$. The critical cooperativity at which bistability starts to occur is somewhat increased. The non-radiative decay provides an additional energy escape channel, such that the trapped incoherent radiation is not strong enough to keep the atoms in the excited state.

Thus we can conclude, that radiative atom-atom interactions do not destroy intrinsic bistability in driven two-level systems, if non-radiative decay is present. This is different from our previous result [26]. The reason for this discrepancy is, that our previous approach essentially neglected the medium effect on the retarded propagation and was therefore inconsistent for larger densities. This would correspond to replacing the retarded propagator of the *interacting* field in Eq.(45) and Fig. 3 by the corresponding free-space Greensfunction.

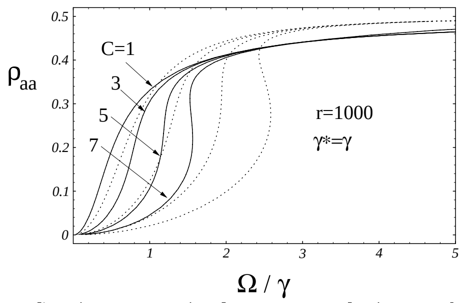

FIG. 12. Stationary excited state population as function of driving-field Rabi-frequency Ω in the presence of additional nonradiative decay ($\gamma^* = \gamma$) and with radiative atom-atom interactions. $r = 1000$. Dotted curves show corresponding behavior without radiative atom-atom interactions for $\gamma^* = \gamma$.

We note that we did not intend to present a comprehensive discussion of effects that could affect intrinsic optical bistability. In particular in atomic vapors collisions may have a much more pronounced effect. Furthermore the depletion of the pump field needs to be taken into account and realistic experimental schemes such as selective reflection spectroscopy [27] need to be considered.

VI. SUMMARY

We have shown that the interaction of a classical radiation field with a dense ensemble of atoms can be described by modified Maxwell-Bloch equations in the Markov-limit. While the equations for the macroscopic classical field, the Maxwell equations, remain unchanged, the atomic equations of motion have additional nonlinear and spatially nonlocal terms that result from the exchange of spontaneous photons between the atoms. The first nonlinear term, the LL-correction, is only present if there is initial coherence or an external coherent field. The nonlinear and nonlocal collective decay and level-shift terms are present whenever there is excited-state population. In the Markov-limit of spectrally broad incoherent radiation inside the medium, the modified Bloch equations have the form of single-atom density matrix equations. They are of the Lindblad type and thus fulfill the formal requirements for conservation of probability and positivity. The effect of the collective decay has been discussed for the examples of an initially excited system of inhomogeneously broadened two-level atoms and intrinsic optical bistability. In the first case we find accelerated decay (superluminescence) in the initial phase and radiation trapping in the final phase. In the long-time limit, where the excited state population is small, the equations of motion coincide with the Holstein equations of radiation trapping [13]. The collective decay modifies intrinsic optical bistability in a strongly driven two-level system. As opposed to our previous prediction [26], bistability persists however, if also non-radiative decay is present. Collective decay and pump processes as well as light shifts are relevant for the population dynamics and are particularly important for ground state coherences in multi-level systems. A detailed discussion of coherence effects in dense multilevel systems as well as the study of cooperative decay processes will be the subject of future work.

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Appendix A: Free-space decay rate

In Eq.(21) we have given the spontaneous decay rate in the atomic medium. We now show that this expression leads to the well-known Wigner-Weisskopf result for the radiative decay of a two-level atom if the interacting field is replaced by the free field. For a single two-level transition with dipole moment along $\vec{e}_\mu$ we have according to (21)

$$\gamma = \frac{\wp^2}{\hbar^2} \int_{-\infty}^{\infty} d\tau \Big\langle [E_{0\,\mu}^+(\vec{r}_0, t+\tau), E_{0\,\mu}^-(\vec{r}_0, t)] \Big\rangle e^{i\omega\tau}. \quad (93)$$

The free-field commutator is given by [22]

$$[E_{0\,\mu}^+(\vec{r}_0, t+\tau), E_{0\,\mu}^-(\vec{r}_0, t)] = \quad (94)$$
$$\frac{\hbar c}{2\epsilon_0} \frac{1}{(2\pi)^3} \int d^3\vec{k}\, k \left(1 - \frac{k_\mu^2}{k^2}\right) e^{-ick\tau}.$$

Substituting (94) into (93) yields

$$\gamma = \frac{\wp^2}{\hbar^2}\frac{\hbar\omega^3}{2\epsilon_0 c^3}\frac{1}{(2\pi)^2}\int d^2\Omega_k \left(1 - \frac{k_\mu^2}{k^2}\right) \tag{95}$$

Finally carrying out the angle-integration leads to

$$\gamma = \frac{\wp^2\omega^3}{3\pi\hbar\epsilon_0 c^3} = \frac{8\pi^2\wp^2}{3\hbar\epsilon_0\lambda^3}, \tag{96}$$

which is the free-space spontaneous emission rate from the Wigner-Weisskopf theory [25].

Appendix B: Dyson equation for real-time Greensfunctions

In this Appendix we derive the integral equations (45) and (46) for the *real-time* GF from the Dyson equation (42) for the *contour* GF. Noting that $D_0^{-+} = 0$ in RWA, we immediately find from (42)

$$\begin{aligned} D^{++} &= D_0^{++} - D_0^{++}\,\Pi^{++}\,D^{++} + D_0^{++}\,\Pi^{+-}\,D^{-+}, \quad (97)\\ &\quad -D_0^{+-}\,\Pi^{--}\,D^{-+} + D_0^{+-}\,\Pi^{-+}\,D^{++},\\ D^{-+} &= \quad -D_0^{--}\,\Pi^{--}\,D^{-+} + D_0^{--}\,\Pi^{-+}\,D^{++}, \quad (98)\end{aligned}$$

where we used a short notation $D_0^{--}\,\Pi^{--}\,D^{-+} = \iint d3d4\, D_{0\,\alpha\mu}^{--}(1,3)\,\Pi_{\mu\nu}^{--}(3,4)\,D_{\nu\beta}^{-+}(4,2)$ and the integration goes over physical times from $-\infty$ to ∞ and over the volume of the sample. Note the sign changes in Eqs.(98) and (97) resulting from the fact that the contour integration on the lower branch goes in the reverse direction. Making use of (38-41) we obtain

$$\begin{aligned} D^{++} &= D_0^{\rm adv} - D_0^{\rm adv}\Big[\Big(\Pi^{++} - \Pi^{-+}\Big)D^{++} - \\ &\Big(\Pi^{+-} - \Pi^{--}\Big)D^{-+}\Big] - D_0^{\rm ret}\Big[\Pi^{-+}\,D^{++} - \Pi^{--}\,D^{-+}\Big], \quad (99)\\ D^{-+} &= -D_0^{\rm ret}\Big[\Pi^{-+}\,D^{++} - \Pi^{--}\,D^{-+}\Big]. \quad (100)\end{aligned}$$

Applying the relation

$$\begin{aligned}\Pi^{\rm adv}(1,2) \equiv \Pi^{++}(1,2) - \Pi^{-+}(1,2) &= \\ \Pi^{+-}(1,2) - \Pi^{--}(1,2)&\end{aligned} \tag{101}$$

and subtracting Eqs.(100) from (99) yields the Dyson equation for $D^{\rm adv} = D^{++} - D^{-+}$:

$$\begin{aligned} D_{\alpha\beta}^{\rm adv}(1,2) &= D_{0\,\alpha\beta}^{\rm adv}(1,2) - \\ &\iint d3\,d4\,D_{0\,\alpha\mu}^{\rm adv}(1,3)\,\Pi_{\mu\nu}^{\rm adv}(3,4)\,D_{\nu\beta}^{\rm adv}(4,2)\end{aligned} \tag{102}$$

where we have restored full notation. Since $D_{\mu\nu}^{\rm ret}(1,2) = D_{\nu\mu}^{\rm adv}(2,1)$ one immediately obtains the corresponding Dyson equation for the retarded propagator

$$\begin{aligned} D_{\alpha\beta}^{\rm ret}(1,2) &= D_{0\,\alpha\beta}^{\rm ret}(1,2) - \\ &\iint d3\,d4\,D_{0\,\alpha\mu}^{\rm ret}(1,3)\,\Pi_{\mu\nu}^{\rm ret}(3,4)\,D_{\nu\beta}^{\rm ret}(4,2)\end{aligned} \tag{103}$$

with

$$\begin{aligned} &\Pi_{\mu\nu}^{\rm ret}(1,2) = \Pi_{\mu\nu}^{++}(1,2) - \Pi_{\mu\nu}^{+-}(1,2) = \\ &\Pi_{\mu\nu}^{-+}(1,2) - \Pi_{\mu\nu}^{--}(1,2) = \\ &\frac{\wp_\mu\wp_\nu}{\hbar^2}\Theta(t_1 - t_2) \\ &\qquad \sum_j \Big\langle \big[\sigma_{j\mu}^\dagger(t_1), \sigma_{j\nu}(t_2)\big]\Big\rangle\,\delta(\vec r_1 \vec r_j)\,\delta(\vec r_2 - \vec r_j).\end{aligned} \tag{104}$$

Thus we obtain the Dyson-equation (46) for the retarded Propagator inside the medium of Sec.IV.

We now turn to D^{-+}. Substituting $\Pi^{--} = \Pi^{-+} - \Pi^{\rm ret}$ in (100) we find

$$D^{-+} = -D_0^{\rm ret}\Pi^{\rm ret}D^{-+} - D_0^{\rm ret}\Pi^{-+}D^{\rm adv}. \tag{105}$$

Iteration of this equations yields

$$\begin{aligned} D^{-+} = -\Big[&D_0^{\rm ret} - D_0^{\rm ret}\Pi^{\rm ret}D_0^{\rm ret} + \quad (106)\\ &D_0^{\rm ret}\Pi^{\rm ret}D_0^{\rm ret}\Pi^{\rm ret}D_0^{\rm ret} - + \cdots\Big]\,\Pi^{-+}D^{\rm adv}\end{aligned}$$

which can be rewritten in the compact form

$$D_{\alpha\beta}^{-+}(1,2) = -\iint d3\,d4\,D_{\alpha\mu}^{\rm ret}(1,3)\,\Pi_{\mu\nu}^{\rm s}(3,4)\,D_{\nu\beta}^{\rm adv}(4,2), \tag{107}$$

where

$$\begin{aligned} &\Pi_{\mu\nu}^{\rm s}(1,2) \equiv \Pi_{\mu\nu}^{-+}(1,2) = \\ &\frac{\wp_\mu\wp_\nu}{\hbar^2}\sum_j \Big\langle\Big\langle \sigma_{j\mu}^\dagger(t_1)\sigma_{j\nu}(t_2)\Big\rangle\Big\rangle\,\delta(\vec r_1 - \vec r_j)\,\delta(\vec r_2 - \vec r_j).\end{aligned} \tag{108}$$

Thus we arrive at Eq.(45) of Sec.IV.

Appendix C: Solution of the Dyson equation for $\widetilde{D}^{\rm ret}$

With the approximations made in Sec.IV-3 we derived the matrix solution (55) for the Fourier-transform of the retarded GF in the medium.

$$\widetilde{\widetilde{\mathbf{D}}}^{\rm ret}(\vec q,\omega,t) = \Big[\mathbf{1} + \widetilde{\widetilde{\mathbf{D}}}_0^{\rm ret}(\vec q,\omega)\cdot\widetilde{\mathbf{P}}^{\rm ret}(\omega,t)\Big]^{-1}\cdot\widetilde{\widetilde{\mathbf{D}}}_0^{\rm ret}(\vec q,\omega). \tag{109}$$

To evaluate this expression we first approximate the Fourier-transform of the free-space retarded propagator. According to [22]

$$\begin{aligned} &D_{0\,\alpha\beta}^{\rm ret}(1,2) = \\ &\frac{i\hbar}{4\pi\epsilon_0 c}\Theta(\tau)\left[\delta_{\alpha\beta}\frac{\partial^2}{\partial\tau^2} - c^2\frac{\partial^2}{\partial x_2^\alpha\partial x_2^\beta}\right]\frac{\delta(r - c\tau)}{r},\end{aligned} \tag{110}$$

with $r = |\vec{x}| = |\vec{r}_1 - \vec{r}_2|$ and $\tau = t_1 - t_2$. Thus

$$\widetilde{D}^{\mathrm{ret}}_{0\,\alpha\beta}(\vec{x},\omega) = -\frac{i\hbar}{4\pi\epsilon_0}\left(\frac{\omega^2}{c^2}\delta_{\alpha\beta} + \frac{x_\alpha x_\beta}{r^2}\frac{\partial^2}{\partial r^2}\right)\frac{e^{-i\omega r/c}}{r}. \tag{111}$$

For large ω, such that $\lambda \ll r$, only the spatial derivative of the exponential contributes and we find

$$\widetilde{D}^{\mathrm{ret}}_{0\,\alpha\beta}(\vec{x},\omega) = -\frac{i\hbar\omega^2}{4\pi\epsilon_0 c^2}\left(\delta_{\alpha\beta} - \frac{x_\alpha x_\beta}{r^2}\right)\frac{e^{-i\omega r/c}}{r}. \tag{112}$$

We now approximate (112) by ignoring the polarization, i.e. by performing an orientation average.

$$\frac{x_\alpha x_\beta}{r^2} \longrightarrow \left\langle \frac{x_\alpha x_\beta}{r^2}\right\rangle = \frac{1}{3}\,\delta_{\alpha\beta}. \tag{113}$$

This approximation is exact when the medium is randomly polarized. (113) leads to

$$\widetilde{D}^{\mathrm{ret}}_{0}(\vec{x},\omega) = -\frac{i\hbar\omega^2}{6\pi\epsilon_0 c^2}\frac{e^{-i\omega r/c}}{r} \tag{114}$$

and thus

$$\widetilde{\widetilde{D}}^{\mathrm{ret}}_{0}(\vec{q},\omega) = -\frac{2i\hbar\omega^2}{3\epsilon_0 c^2}\frac{1}{q^2 - \frac{\omega^2}{c^2} + 2i\epsilon\frac{\omega}{c}}, \tag{115}$$

where we have introduced a small positive constant ϵ to move the pole at $q = \omega/c$ into the lower half of the complex plane.

Substituting (115) into (109) we find

$$\begin{aligned}\widetilde{D}^{\mathrm{ret}}(\vec{x},\omega) &= \frac{1}{(2\pi)^3}\int d^3\vec{q}\;\widetilde{\widetilde{D}}^{\mathrm{ret}}(\vec{q},\omega)\,e^{-i\vec{q}\cdot\vec{x}} \qquad (116)\\ &= -\frac{\hbar\omega^2}{6\pi^2\epsilon_0 c^2}\frac{\delta_{\alpha\beta}}{r}\int_{-\infty}^{\infty}dq \\ &\qquad \frac{q\,e^{-iqr}}{q^2 - \frac{\omega^2}{c^2}\left[1 + \frac{2i\hbar}{3\epsilon_0}\widetilde{P}^{\mathrm{ret}}(\omega)\right] + 2i\epsilon\frac{\omega}{c}}.\end{aligned}$$

We evaluate this integral by contour integration along the real q-axis and back in the lower half plane (note that $r > 0$). In free space ($P = 0$) and for $\epsilon = 0$ we would have two poles on the real axis at $q = \pm\omega/c$. In the free-space case we had to introduce the constant $\epsilon > 0$ to move the pole at $q = \omega/c$ into the lower half-plane (and at the same time to move the pole at $q = -\omega/c$ into the upper one). This is necessary to have retarded propagation. For $\epsilon < 0$ one would obtain the advanced propagator. If an *absorbing* medium is present, $\mathrm{Re}[P^{\mathrm{ret}}]$ is negative and hence adds to ϵ. Therefore again the only contributing pole is that at $q \approx \omega/c$. However, if the medium is *amplifying* there is a problem. In this case $\mathrm{Re}[P^{\mathrm{ret}}]$ is positive and counteracts ϵ. In such a case the pole at $q = -\omega/c$ could move into the lower half-plane and we would obtain an advanced instead of a retarded propagator. The origin of this problem is, that the retarded propagator in an amplifying medium is strictly speaking not Fourier-transformable, since it is an exponentially growing function of distance. In such a case one has to take into account the finite spatial dimensions of the amplifying medium and introduce a cut-off function which leads to a finite-space Fourier-transform. Although this lacks mathematical rigor, we now assume that the effect of the cut-off function is modeled by a sufficiently large value of ϵ, such that the pole at $q \approx \omega/c$ remains in the lower half-plane. With this we find

$$\widetilde{D}^{\mathrm{ret}}(\vec{x},\omega) = -\frac{i\hbar\omega^2}{6\pi\epsilon_0 c^2}\frac{e^{q_0'' r}}{r}\,e^{-iq_0' r}, \tag{117}$$

where

$$q_0(\vec{r},\omega,t) = \frac{\omega}{c}\left[1 + \frac{i\hbar}{3\epsilon_0}\widetilde{P}^{\mathrm{ret}}(\vec{r},\omega,t)\right]. \tag{118}$$

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