Modification of local field effects in two level systems due to quantum corrections

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Abstract: Quantum corrections to the Lorentz–Lorenz formula are given for a dense ensemble of atoms interacting with the quantized radiation field. The influence of these corrections on local-field effects in two-level systems is discussed in the non-cooperative limit. For initially inverted atoms we find superluminescence and radiation trapping. Furthermore it is shown that the quantum corrections set strong limitations to intrinsic optical bistability.

OCIS codes: (190.1450) Bistability; (020.1670) Coherent optical effects; (270.2500) Fluctuations, relaxation and noise

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1. Introduction

The interaction of an ensemble of atoms with the radiation field 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 Lorentz and Lorenz [1, 2] it is known that the classical electric field, that couples to an oscillator in a dense medium differs from the macroscopic (Maxwell) field by a term proportional to the medium polarization. If the medium (or the host material in compound systems) has a small absorption, but a large index of refraction, the local field can be substantially different from the Maxwell field [3, 4, 5]. A number of interesting phenomena result from this effect. For example shifts of atomic absorption lines have been observed [6, 7], nonlinear susceptibilities can be enhanced [8], and mirrorless (i.e. intrinsic) optical bistability [4, 9, 10] and piezo-photonic switching from amplification to absorption [11] have been predicted in resonantly driven systems.

On the other hand, when the atomic density becomes large, the quantum nature of the field is of increasing importance. In his famous work [12] Dicke showed that a large number of excited atoms within a cubic wavelength emit photons in a cooperative way. This phenomenon called superfluorescence has been extensively studied since then [13, 14]. But even if the system does not fulfill the conditions for cooperative evolution (due to e.g. inhomogenous broadening), the presence of spontaneous photons can not be neglected. It is known for example that the amplification of spontaneously emitted radiation can strongly affect the dynamics of a dense excited medium.

Starting from a fully quantized interaction model we have derived in Ref.[15] a single-atom density-matrix equation in the non-cooperative limit, i.e. when atom-atom correlations can be neglected. This nonlinear and nonlocal density-matrix equation contains the semiclassical Lorentz-Lorenz local field as well as the dominant quantum corrections which lead to additional relaxation terms and level shifts. We here apply this equation to the case of a dense ensemble of two-level atoms.

In the first part of the paper we discuss the spontaneous decay of an initially inverted sample of atoms in a thin cylindrical slab. We find, that the radiative decay is strongly accelerated in the initial phase due to the atom-atom interaction via the quantized field, thus describing superluminescence. For larger times the decay is however slowed down. The reabsorption of spontaneously emitted photons prevents the radiation energy from escaping, a phenomenon known as radiation trapping. Both effects, superluminescence and radiation trapping are here obtained in the framework of a nonlinear density matrix equation derived from a first-principles calculation.

In the second part of the paper we discuss the influence of the quantum terms on intrinsic optical bistability in a resonantly driven, dense ensemble of two-level atoms in a spherical geometry. The nonlinearity of the Bloch equation without quantum corrections - caused by the dependence of the Lorentz-Lorenz local field on the atomic polarization - can lead to bistability even without external feedback [9, 10]. It is shown here that the superluminescence field destroys this bistability to a large extend.

2. One-atom master equation: Lorentz-Lorenz local field and quantum corrections

The interaction between atoms and field can be consistently described by *single-atom* Bloch-type equations only if atom-atom correlations are neglected, i.e. in the non-cooperative limit. This approximation is justified in highly symmetric geometries or

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if the inverse of the so-called superradiance time corresponding to a certain transition

$$T_R^{-1} \equiv N_e \gamma \mu \tag{1}$$

is smaller than the respective inhomogeneous width [13]. Here N_e denotes the number of excited atoms and μ is a geometry factor, which for a pencil-shaped medium of radius R scales as $3\lambda^2/8\pi R^2$. Throughout the present paper we will assume that this condition is fulfilled. As shown in Ref.[15] each atom sees in this case a radiation field with a coherent component given by the Lorentz-Lorenz formula, i.e. the Maxwell field plus a term proportional to the mean polarization of the probe atom. In addition there is an incoherent field component which depends on single-atom correlation functions of the surrounding atoms. When this incoherent component is spectrally broad, one can derive (nonlinear and nonlocal) single-atom density matrix equations. For a two-level

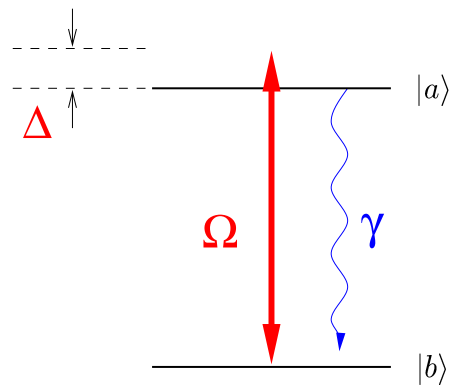

Fig.1: Two level system with decay γ and cw coherent field Ω.

system as shown in Fig. 1 with inversion $\rho_{aa} - \rho_{bb} = w$ and polarization ρ_{ab} driven by a coherent field of Rabi-frequency Ω they read

$$\begin{aligned} \dot{w}^{(l)} &= -(\gamma + 2g_l)(w^{(l)} - w_0^{(l)}) - 2\mathrm{i}\Omega \left(\rho_{ab}^{(l)} - c.c.\right) \\ \dot{\rho}_{ab}^{(l)} &= -\left[\frac{\gamma}{2} + g_l + \mathrm{i}(\Delta + h_l)\right] \rho_{ab}^{(l)} - \mathrm{i}\Omega_{lL} w^{(l)} \end{aligned} \tag{2}$$

where the superscript l denotes the lth atom, γ is the radiative decay rate from $|a\rangle$ to $|b\rangle$ and $\Delta = \omega_0 - \nu$ the detuning between the atomic and field frequencies. Due to the Lorentz–Lorenz terms the atomic polarization $\rho_{ab}^{(l)}$ is not driven by the Maxwell-field but by the local field with Rabi-frequency

$$\Omega_{lL} = \Omega + \mathcal{C}\gamma\rho_{ab}^{(l)}. \tag{3}$$

The parameter

$$\mathcal{C} = \frac{\lambda^3 \mathcal{N}}{4\pi^2}, \tag{4}$$

where $\mathcal{N}$ is the atomic density and λ the transition wavelength, is called cooperativity parameter.

$$w_0^{(l)} = -\frac{\gamma}{\gamma + 2g_l} \tag{5}$$

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is the equilibrium inversion, which approaches -1 for $g_l \to 0$. The quantum corrections are described by the quantities h_l and g_l:

$$
\begin{aligned}
g_l &= -2\,\mathrm{Re}\,[s_l]\ , \\
h_l &= \ \ 2\,\mathrm{Im}\,[s_l]\ , \\
s_l(t_1) &= \frac{\wp^4}{\hbar^4}\sum_j \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_1} dt_3 \int_{-\infty}^{t_1} dt_4\ \mathrm{e}^{\mathrm{i}\omega_0((t_1-t_2)-(t_3-t_4))} \times \\
&\quad \times D^{ret}(\vec{r}_l, t_1; \vec{r}_j, t_3) D^{ret}(\vec{r}_l, t_2; \vec{r}_j, t_4) \langle\langle \sigma_j^-(t_4)\sigma_j^+(t_3)\rangle\rangle\ .
\end{aligned} \tag{6}
$$

Here $\sigma_j^+ = |b\rangle_{jj}\langle a|$ denotes the positive frequency part of the atomic dipole operator in a frame rotating with the frequency of the atomic transition. We assume that the atomic dipoles are oriented in the z-direction. $\langle\langle xy\rangle\rangle \equiv \langle xy\rangle - \langle x\rangle\langle y\rangle$, and D^{ret} is the retarded Greens function of the electromagnetic field

$$
D^{ret}(\vec{r}_1, t_1; \vec{r}_2, t_2) = \frac{\mathrm{i}\hbar}{4\pi\epsilon_0 c}\Theta(\tau)\left[\frac{\partial^2}{\partial\tau^2} - c^2\cos^2\vartheta\,\frac{\partial^2}{\partial r^2}\right]\frac{\delta(r-c\tau)}{r}\ , \tag{7}
$$

with $\tau = t_1 - t_2$, $r = |\vec{r}_1 - \vec{r}_2|$, and the angle between $\vec{r}$ and the z-axis ϑ.

As can be seen from Eq. (2), g_l assumes in a two-level medium the form of an additional incoherent pumping term, and h_l the form of an additional detuning. As will be seen later, h_l is, in a two-level system, itself proportional to the detuning Δ.

Using the quantum regression theorem [16], one can calculate the atomic cumulants $\langle\langle\sigma_j^-\sigma_j^+\rangle\rangle$ from the Heisenberg-Langevins equation for σ_j^- (σ_j^+) and $\sigma_{aa}^j = |a\rangle_{jj}\langle a|$, which are formally equivalent to the density matrix equations. Under adiabatic conditions this then yields the coefficients g_l and h_l in terms of the atomic polarizations $\rho_{ab}^{(j)}$ and inversions $w^{(j)}$, which have to be calculated in a selfconsistent way.

In contrast to the Lorentz-Lorenz term, which is a contact contribution and thus depends only on the polarization of the probe atom itself, the quantum corrections depend on the polarization and inversion of all other atoms. Hence one has to take into account the spatial degrees of freedom, which makes the calculation considerably more difficult. If we however assume that the sample remains homogeneous, given that the initial condition and/or the external driving field is homogeneous, we can carry out the summation over the atomic indices (or in continuum approximation the spatial integration) in Eq.(6) explicitly. We will use this approximation in the following but will also remark on its limitations.

3. Physical consequences

In this section we discuss the physical consequences of the nonlinear contributions in the density matrix equation (2) for two examples. The first one is an ensemble of initially excited, inhomogeneously broadened two-level atoms in a pencil-like configuration. The second one is an ensemble of two-level atoms in a sphere of radius R irradiated by a cw coherent field. In this system intrinsic optical bistability was predicted by a semiclassical model where only the Lorentz-Lorenz local field was taken into account [9, 10].

3.1 Superluminescence and radiation trapping

A gas of two-level atoms in a pencil-shaped volume is well-known to show superluminescence (amplified spontaneous emission) for higher, and superradiance for very high densities. As noted before we here restrict ourselves to the first case and neglect atom-atom correlations and the associated cooperative effects. As an initial condition we assume that all population is in the upper level $|a\rangle$. In this case we have just one equation of motion

$$\dot{\rho}_{aa} = -(\gamma + 2g)\rho_{aa} + g. \tag{8}$$

The effective decay rate g can be calculated in a selfconsistent way if $\langle\langle\sigma^-(t-\tau)\sigma^+(t)\rangle\rangle$ can be considered quasi–stationary. This is the case, for example, if the medium is inhomogenously broadenend. After some algebra we eventually arrive at

$$\frac{g}{\gamma} = \eta \frac{1 + 2\frac{g}{\gamma}}{1 + \left(\frac{\gamma}{\Delta_D}\right)^2 \left(\frac{1}{2} + \frac{g}{\gamma}\right)^2} \rho_{aa} \,, \tag{9}$$

$$\eta = \left(\frac{3}{4}\right)^3 \mathcal{N}\lambda^2 d \left(\frac{\gamma}{\Delta_D}\right)^2 = \frac{27\pi^2}{16}\mathcal{C}\frac{d}{\lambda}\left(\frac{\gamma}{\Delta_D}\right)^2$$

for an atom on the cylinder axis with diameter d where Δ_D is the inhomogeneous width. The quantum part adding to the detuning vanishes in this case, i.e. $h = 0$. Eq. (8) can easily be integrated numerically. The corresponding results for the upper level population ρ_{aa} is plotted in Fig. 2 for different density parameters η. One clearly

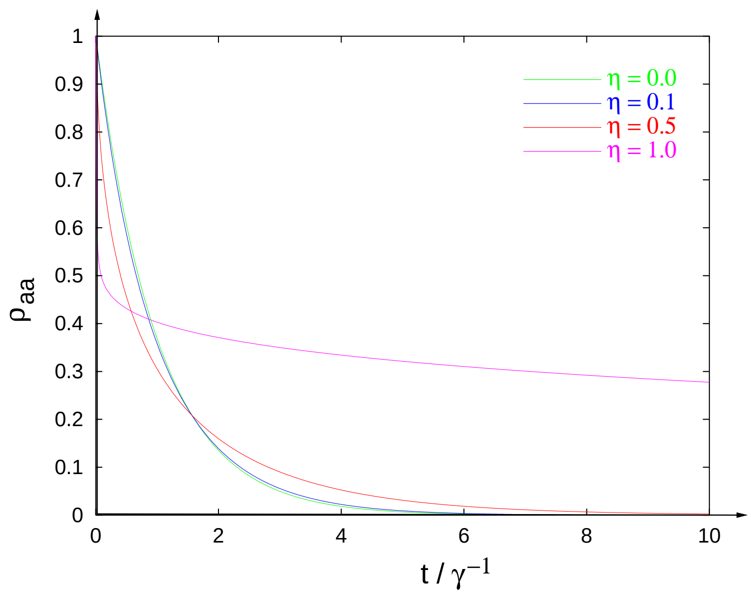

Fig.2: The decay of the the upper-level population ρ_{aa} for a ratio of the radiative to the inhomogeneous linewidth $\gamma/\Delta_D = 0.1$ including quantum corrections for different values of η.

recognizes an accelerated decay for small times which is a manifestation of amplified spontaneous emission or superluminescence. For longer times, the decay rate of the population slows down substantially. This is due to the large intensity of incoherent radiation in the sample which partially reexcites the atoms. Due to this reabsorption, the radiation energy is trapped inside the medium. It is notable that the transition between accelerated and decelerated spontaneous emission happens exactly at $\rho_{aa} = 0.5$. This result could be expected, since the additional quantum term has exactly the form of an incoherent pump. Such a field always tries to equalize the population in the two levels.

It should be noted that for values of $\eta > \eta_0 = 1 + (\gamma/2\Delta_D)^2$ there is, except

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for ${\rho_{aa}}^{ss(1)} = 0$ a second stationary solution of Eq.(8).:

$$ {\rho_{aa}}^{ss(2)} \to \frac{1}{2} - \frac{\gamma}{4\Delta_D\sqrt{\eta - 1}}. \tag{10} $$

This slightly unphysical long-time behaviour results from the assumption of spatial homogeneity, which breaks down in this case. When the absorption length of the medium, $[\mathcal{N}\lambda^2(-w)]^{-1}$, becomes sufficiently small, i.e. comparable to the dimension of the medium, the spatial dependence needs to be taken into account. Starting from a homogeneous atomic inversion the intensity of the superluminescence radiation is at the surface smaller as compared to the center. Hence the incoherent radiation gets trapped in a region with effective radius $d_{\text{eff}} < d$ which shrinks in time. Thus η decreases and eventually becomes smaller than the critical value η_0. A proper treatment of the collective decay for larger values of η clearly requires to include the spatial degrees of freedom. This however goes beyond the scope of the present communication and will be discussed in a future publication.

3.2 Limitations to intrinsic bistability

The usual thin-medium Bloch equations for the atomic evolution in an external radiation field are linear and therefore permit only a single, well-defined solution. With a feedback mechanism, however, the effective evolution becomes nonlinear and optical bistability is possible. An interesting feature of the semiclassical Bloch equations including the Lorentz-Lorenz local field is the explicit nonlinearity (see Eqs.(2) and (3)). When this nonlinearity is sufficiently large optical bistability is possible even without external feedback [9, 10]. Since the required atomic density is extremely high, it is conceivable that quantum corrections can substantially change the predicted behaviour. We therefore analyze here the effect of quantum corrections to the Lorentz-Lorenz formula on intrinsic optical bistability of driven two-level atoms in a spherical sample of radius R.

The equations of motion for a two-level system with an additional cw coherent field detuned by Δ from the atomic resonance, are given by Eqs. (2). In steady state the quantum corrections can be calculated in a straightforward (but somewhat lengthy) way. For an atom in the center of the sphere they are given by

$$ \begin{aligned} g &= \frac{24\pi}{5}\mathcal{C}\varrho\gamma^2 \Bigg[\frac{\gamma + 2g}{|\Gamma|^2}\left(\rho_{aa} - |\rho_{ab}|^2\right) - \frac{|\Omega_L|^2\left((\gamma/2+g)^2 - (\Delta+h)^2\right)}{|\Gamma|^4\left(\alpha+\gamma/2+g\right)}\rho_{aa} \\ &\quad + \frac{\mathrm{i}}{\alpha+\gamma/2+g}\left(\frac{\Omega_L\rho_{ba}}{\Gamma} - \frac{\Omega_L^*\rho_{ab}}{\Gamma^*}\right)\left(\rho_{aa} - \mathrm{i}\left(\frac{\Omega_L^*\rho_{ab}}{\Gamma} - \frac{\Omega_L\rho_{ba}}{\Gamma^*}\right)\right)\Bigg], \\ h &= \frac{24\pi}{5}\mathcal{C}\varrho\gamma^2 \Bigg[-2\frac{\Delta+h}{|\Gamma|^2}\left(\rho_{aa} - |\rho_{ab}|^2\right) + \frac{2|\Omega_L|^2\left(\gamma/2+g\right)\left(\Delta+h\right)}{|\Gamma|^4\left(\alpha+\gamma/2+g\right)}\rho_{aa} \\ &\quad + \frac{1}{\alpha+\gamma/2+g}\left(\frac{\Omega_L\rho_{ba}}{\Gamma} + \frac{\Omega_L^*\rho_{ab}}{\Gamma^*}\right)\left(\rho_{aa} - \mathrm{i}\left(\frac{\Omega_L^*\rho_{ab}}{\Gamma} - \frac{\Omega_L\rho_{ba}}{\Gamma^*}\right)\right)\Bigg], \end{aligned} \tag{11} $$

where $\varrho = R/\lambda$ and

$$ \begin{aligned} \Gamma &= (\frac{\gamma}{2} + g) + \mathrm{i}(\Delta + h), \\ \alpha &= \frac{(\gamma+2g)|\Omega_L|^2}{|\Gamma|^2}. \end{aligned} $$

Formally, $\varrho = 0$ corresponds to the semiclassical limit including local field terms but without quantum corrections. In this case the steady-state density matrix equations

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reduce to a single cubic equation for the excited state population. In Fig. 3 we have plotted the solution for ρ_{aa} as a function of the Rabi-frequency Ω for $\Delta = 0$ and different values of $\mathcal{C}$. As can be seen, for $\mathcal{C} > 3\sqrt{3}/2 \approx 2.598$ the system becomes bistable.

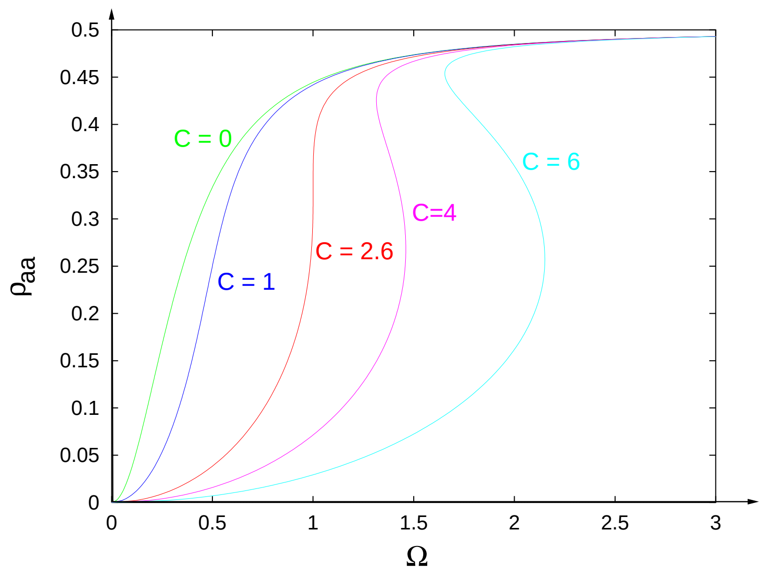
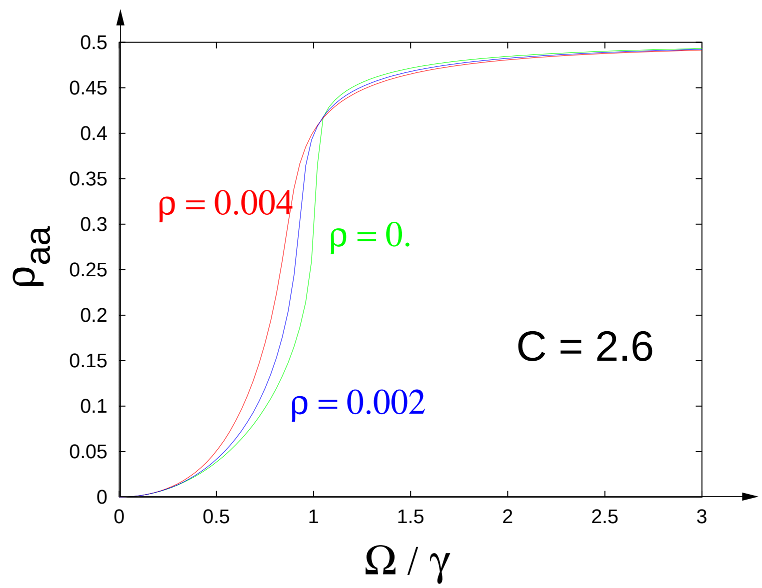


Fig.3: The upper-level population ρ_{aa} plotted as function of the driving-field Rabi frequency Ω. Quantum corrections are not taken into account. Different curves correspond to different cooperativity $\mathcal{C}$. The transition between normal and bistable regime occurs at $\mathcal{C} \sim 2.6$.

For $\varrho > 0$ the set of equations gets more involved and can only be solved numerically. On resonance where for $\varrho = 0$ the bistability is strongest we again have $h = 0$. The additional term proportional to g acts exactly like an additional incoherent pump on the system, and it can be expected that already for very low values of ϱ the bistablity is diminished. In Fig. 4 we show the influence of the quantum terms for $\mathcal{C} = 2.6$ (Fig. 4a) and for $\mathcal{C} = 4$ (Fig. 4b) for increasing values of ϱ. The stationary solutions were obtained by numerically integrating the nonlinear Bloch equations and considering the long-time limit for different initial conditions. In this way only the stable stationary solutions were obtained (uppermost and lowermost parts of the curves). The shaded areas illustrate regions of bistability. We note that already for a radius on the order of a fraction of the wavelength the incoherent field can destroy bistability completely. The physical origin of this effect can be explained as follows: Already for very small radii the number of excited atoms within the sample is large enough for the considered densities such that the incoherent power $P_{\rm inc}$ becomes comparable to the coherent one $P_{\rm coh}$. Since

$$\frac{P_{\rm inc}}{P_{\rm coh}} \sim 8\pi C \varrho \left(\frac{\gamma}{\Omega}\right)^2, \tag{12}$$

the effect becomes even more pronounced when the cooperativity parameter $\mathcal{C}$ is increased. Although spatial inhomogeneity has again been neglected here, it thus seems questionable that intrinsic bistability can be observed in driven two-level systems with purely radiative interaction. If, on the other hand, other interaction mechanisms are present as well, as in the experiment by Rand et al. [17], bistability can be observed. In Ref. [17], the presence of pair upconversion substantially enhanced the hysteresis behaviour. With that also the region of Rabi-frequencies for which bistability exists



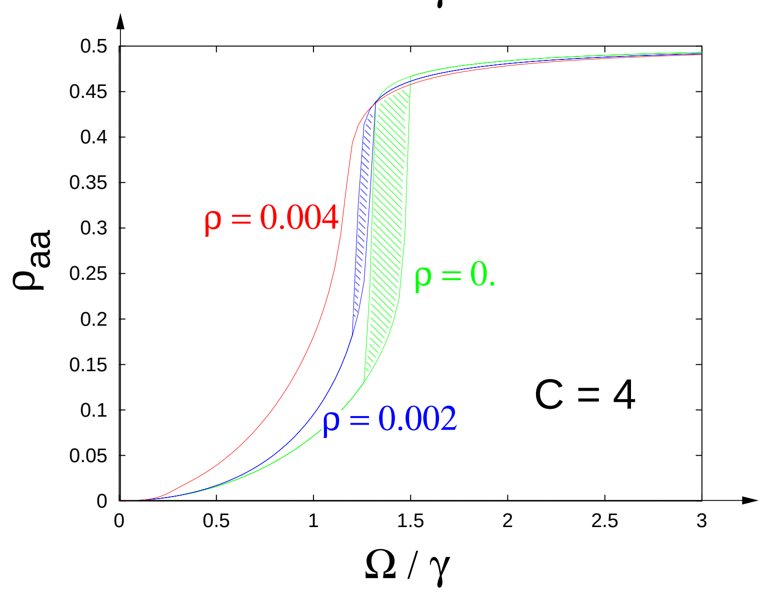


Fig.4: The curves of the previous figure, but including quantum corrections for $\mathcal{C} = 2.6$ (a) and $\mathcal{C} = 4$ (b). The parameter ρ assumes the values 0, 0.002, 0.004.

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was shifted from $\Omega \sim \gamma$ to larger values by orders of magnitude. Consequently $P_{\rm coh}$ is substantially increased and bistability is destroyed only for much larger values of $\varrho = R/\lambda$.

4. Conclusions

We have shown that in a dense medium the radiative interaction between atoms due to spontaneous emission and reabsorption plays an important role and cannot be neglected. While the size of the Lorentz-Lorenz local field term is essentially determined by the cooperativity parameter $\mathcal{C}$, the size of the quantum corrections scales as $\mathcal{C}\varrho\rho_{aa}$. Since $\varrho = R/\lambda$ (where R is a typical linear dimension of the sample) is usually much larger than unity, the quantum corrections very often dominate over the local field terms. As we have shown for the example of intrinsic optical bistability in a purely radiatively interacting system, the quantum corrections can substantially change local field effects if there is an appreciable amount of excited state population. This conclusion does however not rule out intrinsic optical bistability if other interactions are present or in multi-level systems. In fact it has been shown in Ref.[18] that utilizing atomic coherence effects in such systems involving weak transitions, a large index of refraction i.e. large local field effects can be combined with small excited state population. Such systems will be studied in our future work.

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