Correlation in superradiance: A closed-form approach to cooperative effects

S. F. Yelin, 1, 2 M. Koštrun, 1, 2 T. Wang, 1 and M. Fleischhauer 3

1 Department of Physics, University of Connecticut, Storrs, CT 06269
2 ITAMP, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA 02138
3 Universität Kaiserslautern, Kaiserslautern, Germany

(Dated: July 28, 2018)

We have developed a novel method to describe superradiance and related cooperative and collective effects in a closed form. Using the method we derive a two-atom master equation in which any complexity of atomic levels, semiclassical coupling fields and quantum fluctuations in the fields can be included, at least in principle. As an example, we consider the dynamics of an initially inverted two-level system and show how even such in a simple system phenomena such as the initial radiation burst or broadening due to dipole-dipole interactions occur, but it is also possible to estimate the population of the subradiant state during the radiative decay. Finally, we find that correlation only, not entanglement is responsible for superradiance.

I. INTRODUCTION

Following the appearance of Dicke’s paper 1 on spontaneous emission of radiation by an assembly of atoms, cooperative effects such as superradiance were studied by many authors, e.g. 2. Recently, there was a revived interest in, so called, Dicke states because of their potential application in quantum information processing 3, 4, 5, 6 and their importance in the behavior of Bose-Einstein Condensates 7.

The main reasons to revisit superradiance in this article are (a) the need for an improved calculational model to support the revived interest, and (b) a set of novel phenomena that could be discovered using this model, such as subradiance and chirping, (c) the need for a simplified formalism to treat more complicated level systems, and (d) the improved understanding of the phenomena of collective vs. cooperative effects.

We have recently developed a novel method to study collective effects in optically dense media 8. Whereas collective effects are caused by a radiative dipole-dipole interaction between the atoms, cooperative effects also include the exchange of virtual photons between the atoms that leads to the formation of Dicke states. The difficulty of calculating effects including atom-atom cooperation has been the intrinsically large number of interconnected degrees of freedom, even if just a few particles are involved. In this article we incorporate cooperative effects into our formalism, leading to an effective two-atom master equation formalism for superradiance.

From the simplest version of this calculation we are able to obtain good agreement with observed signatures of superradiance including the effects of dissipation and the unique temporal build-up of a sharp flash of radiation. In addition, our new formalism allows more complicated level structures, additional fields, and polarization to be easily considered as well.

What is the main difference between collective and cooperative effects? In this context, we define as “collective effects” all those, where a multi-particle ensemble responds differently than just the sum of all single-particle responses. Examples of collective effects in a atomic gas with radiative interactions include amplified spontaneous emission and radiation trapping (see e.g. 8). “Cooperative effects,” on the other hand, specifically arise from the exchange interaction between any two particles. The best-known example for this exchange interaction is the energy splitting between the symmetric and anti-symmetric two-atom superposition states, \((|ab\rangle \pm |ba\rangle)/\sqrt{2}\), where \(|a\rangle\) and \(|b\rangle\) are the excited and ground states of a single two-level atom. In this article, we will quantify the exchange interaction as the non-diagonal two-atom term of a two-atom density matrix \(\rho\), \(\rho_{ab,ba} = 0\). All typical signatures of superradiant behavior can be explained using this term.

Since the term \(\rho_{ab,ba}\) describes the coherence between any two atoms, we will refer to it as “correlation,” and we will show (see, e.g., Fig. 2) that the correlation is the reason for the build-up of superradiance.

II. MODEL

We consider an arrangement of \(N\) atoms which mutually interact via quantized electric field in dipol and rotating-wave approximations (similar as was done in 8). We are interested in cooperative phenomena of the ensemble so we distinguish two atoms in the arrangement, say 1 and 2, and write the total Hamiltonian as

\[
H = H_{\text{field}} + \sum_{i=1,2} \left\{ H_0^i - \vec{p}_i \left( \vec{E}_i + \vec{E}_i^* \right) \right\} + \sum_{j \neq 1,2} \left\{ H_0^j - \vec{p}_j \left( \vec{E}_j + \vec{E}_j^* \right) \right\},
\]

(1)

for the probe atoms \(i = 1, 2\) and all other atoms \(j \neq 1, 2\). Here, \(H_0^i\) and \(H_{\text{field}}\) are the free Hamiltonian of the \(i\), \(j\)-th atom at location \(\vec{r}_{i,j}\), and that of the quantized radiation field, \(\vec{E}\), respectively. Furthermore, \(\vec{E}_{i,j} = \vec{E}(\vec{r}_{i,j})\).
is the quantized field at the position of the $i, j$th atom, while $\mathcal{E}_j = \mathcal{E}(\vec{r}_j)$ is the external classical driving field. $\hat{p}_{i,j} = \hat{p}(\vec{r}_{i,j})$ is the atom dipole operator.

is the quantized field at the position of the $i, j$th atom, while $\mathcal{E}_j = \mathcal{E}(\vec{r}_j)$ is the external classical driving field. $\hat{p}_{i,j} = \hat{p}(\vec{r}_{i,j})$ is the atom dipole operator.

In the next step we move to an interaction picture using the time evolution operator on the appropriately chosen Schwinger-Keldysh contour $C$, shown in Fig. II

\[ S_C = T_C \exp \left\{ \frac{i}{\hbar} \int_{c_{-}}^{c_{+}} d\tau \sum_{j=1,2} \hat{p}_j \left( \hat{E}_j + \mathcal{E}_j \right) \right\}, \tag{2} \]

where $T_C$ is a time ordering operator on the Schwinger-Keldysh contour, and is identical to standard time ordering operator $T$ on the upper or $+$ branch, and to its inverse $T^{-1}$ on the lower or $-$ branch. By this formal procedure each physical time $t$ is replaced by two times on the contour, $t_+$ when going from $-\infty$ to $+\infty$ on the upper or $+$ branch, and $t_-$ when going in opposite direction on the lower or $-$ branch. In this notation, an infinitesimal complex quantity is added to what was previously real time $\tau$ in order to obtain $\hat{\tau}$, where the sign of the infinitesimal complex part depends on the branch of the Schwinger-Keldysh contour.

We find the expectation value of Eq. (2) by replacing formally

\[ \langle T_C \exp \{ \hat{A} \} \rangle = \exp \left\{ \sum_n \langle \langle T_C \hat{A}^n \rangle \rangle \right\}, \tag{3} \]

where $\hat{A}$ is the operator in the exponent of Eq. (2). The cumulants contain the pure correlation, e.g., $\langle \langle \hat{x} \rangle \rangle \equiv \langle \hat{x} \rangle$, $\langle \langle \hat{x} \hat{y} \rangle \rangle \equiv \langle \hat{x} \rangle \langle \hat{y} \rangle - \langle \hat{x} \rangle \langle \hat{y} \rangle$, etc. Assuming that the radiation field is Gaussian allows us to bring to a tractable form the cumulants involving the radiation field, which appear when averaging the evolution operator $\mathcal{E}_L = \mathcal{E} + \langle \hat{E} \rangle$ for the local field seen by each of the two atoms. The two cumulants that appear in Eq. (3) are

\[ D_{i\mu,j\nu}(\tau_1, \tau_2) = \langle \langle T_C \hat{E}_{i\mu}(\hat{\tau}_1) \hat{E}_{j\nu}(\hat{\tau}_2) \rangle \rangle e^{-i\omega(\tau_1 - \tau_2)} \]

\[ C_{i\mu,j\nu}(\tau_1, \tau_2) = \langle \langle T_C \hat{E}_{i\mu}(\hat{\tau}_1) \hat{E}_{j\nu}(\hat{\tau}_2) \rangle \rangle e^{i\omega(\tau_1 - \tau_2)} \]

The function $D$, for example, can then be expressed as

\[ D_{i\mu,j\nu}(\tau_1, \tau_2) = e^{-i\omega(\tau_1 - \tau_2)} D_{i\mu,j\nu}^{+\pm}, \tag{6} \]

with

\[ \langle \langle T \hat{E}_{i\mu}(\tau_1, \tau_1) \hat{E}_{j\nu}(\tau_2, \tau_2) \rangle \rangle = D_{i\mu,j\nu}^{+\pm}, \]

\[ \langle \langle T^{-1} \hat{E}_{i\mu}(\tau_1, \tau_1) \hat{E}_{j\nu}(\tau_2, \tau_2) \rangle \rangle = D_{i\mu,j\nu}^{-\pm} \]

\[ \langle \langle \hat{E}_{i\mu}(\tau_1, \tau_1) \hat{E}_{j\nu}(\tau_2, \tau_2) \rangle \rangle = D_{i\mu,j\nu}^+ \]

\[ \langle \langle \hat{E}_{i\mu}(\tau_1, \tau_1) \hat{E}_{j\nu}(\tau_2, \tau_2) \rangle \rangle = D_{i\mu,j\nu}^- \]

where we used the properties of the time ordering operator $T_C$ on the Schwinger-Keldysh contour. In the expression above a subscript sign “+” or “-” to the time variable $\tau_{1,2}$ indicates the branch of the Keldysh contour on which the time variable is located. Similar expression can be written down for function $C$ as well. We assume that there is only one type of atoms so we set

\[ \hat{p}_{i\mu}(\vec{r}_i, t) = \hat{p}_{i\mu}(\vec{r}_i, t) + \hat{\sigma}_{i\mu}(t) \]

\[ \hat{p}_{i\mu}(\vec{r}_i, t) = \hat{p}_{i\mu}(\vec{r}_i, t) + \hat{\sigma}_{i\mu}(t) \]

where $\hat{\sigma}_{i\mu}$ is the dipole matrix element for all polarizations, and $\hat{\sigma}(t)$ is the dimensionless dipole vector operator of the individual atom $i$. For real times, the effective time evolution operator $C$ becomes, after all the contributions
are real and imaginary part of the Fourier transformed cumulant $D^{-, \pm}$, and so they are mutually related via the Kramers-Kronig relationship.

Term $\Gamma$ describes the decay and pump rates induced by the incoherent photons inside the medium, while the term $\gamma$ is the spontaneous “down rate” in the atomic medium. Term $H$ describes a collective light shift in addition to the inhomogeneous broadening, and is incorporated in our formalism. Term $\hbar$ is a spontaneous contribution to the light shift, the diagonal terms of which amount to the Lamb shift. As we are not interested in the Lamb shift, we consider these terms either included in the free Hamiltonian $H_0$ (diagonal), or zero (off-diagonal). With the help of Eq. (11), we can write the effective two-atom density matrix equation as

$$\dot{\rho} = -\frac{i}{\hbar}[H_0, \rho] + \frac{i}{\hbar} \sum_{\mu,\nu} \phi_{\mu} \sum_{j=1}^{2} \left[ \sigma_{j\mu} \mathcal{E}_{L,\mu}(\vec{r}_j) + \sigma_{j\mu}^\dagger \mathcal{E}_{L,\mu}(\vec{r}_j) \right] \rho + \frac{i}{\hbar} \sum_{\mu,\nu} \sum_{j=1}^{2} H_{j\mu,j\nu} \left[ [\sigma_{j\mu}, \sigma_{j\nu}^\dagger], \rho \right] - \sum_{\mu,\nu} \sum_{i,j=1}^{2} \frac{\Gamma_{ij,j\mu}}{2} \left( \left[ \rho \sigma_{ij}, \sigma_{j\mu}^\dagger \right] + \left[ \sigma_{ij}, \sigma_{j\mu}^\dagger \rho \right] \right) - \sum_{\mu,\nu} \sum_{i,j=1}^{2} \frac{\Gamma_{ij,j\mu} + \gamma_{ij,j\mu}}{2} \left( \left[ \rho \sigma_{ij}, \sigma_{j\mu} \right] + \left[ \sigma_{ij}^\dagger, \sigma_{ij} \rho \right] \right),$$

where $\mathcal{E}_{L,\mu}(\vec{r}_j)$ is the local (Lorentz-Lorenz) field felt by the $j$-th atom. Eq. (11) is one of the main results of this paper. Up to this point we assumed a homogenous atomic medium, made a Markov approximation, and neglected correlations of higher than second order. All coherent fields are displayed in the term $\mathcal{E}_L$, together with the first-order quantum corrections which take care of the local field. All second-order corrections are in the terms with $H$ and $\Gamma$. This equation looks similar to the effective master equation for large times, derived in Refs. [12, 13]. Note, however, that Eq. (11) is a two-atom equation! In addition, the second-order quantum correction terms, which have quite a different form in our case, contain most of the physics in the context of cooperative effects. It is also possible to find a closed-form expression for them, as will be seen in the next section.

III. COOPERATIVE EFFECTS IN A HOMOGENEOUS GAS OF TWO-LEVEL ATOMS

For a gas of two-level atoms there is only one transition, and the coordinate indices $\mu$ and $\nu$ can be dropped; the effective two-atom two-level master equation (11) as-
sumes the form

\[ \dot{\rho} = -\frac{i}{\hbar} [H_0, \rho] + \sum_{j=1,2} \frac{i}{\hbar} \mathcal{V} \left[ \sigma_j \mathcal{E}_L^y (\vec{r}_j) + \sigma_j^+ \mathcal{E}_L^x (\vec{r}_j), \rho \right] \]

\[ + \frac{i}{\hbar} \sum_{j=1,2} H_{aj,j} \left[ \sigma_j, \sigma_j^+ \right], \rho \]

\[ - \frac{1}{2} \sum_{i,j=1,2} \Gamma_{ij} \left[ \left( \rho \sigma_i \sigma_j^+ + \sigma_i^+ \sigma_j \right) \right] \rho \]

\[ - \frac{1}{2} \sum_{i,j=1,2} \left( \Gamma_{ij} + \gamma_{ij} \right) \left( \left( \rho \sigma_i^+ \sigma_j + \sigma_i \sigma_j^+ \right) \right) \].

Let us introduce the notation for the two-atom density matrix as following,

\[ \rho_{\alpha \beta \gamma \delta} \equiv \langle \alpha_1 \gamma_2 | \rho | \beta_1 \delta_2 \rangle. \] Each of the two atoms can be in the ground state \(|b\rangle\), or the excited state \(|a\rangle\), and \(|\alpha_1, \gamma_2\rangle\) is a product \(|\alpha_1\rangle \cdot |\gamma_2\rangle\) of the states of the first and second atom. The atomic lowering operator \(\sigma\) assumes the form \(\sigma = |b\rangle \langle a|\). This allows us to introduce three real functions, namely \(a\), \(d\) and \(n\), as follows

\[ 2a = 2\rho_{aa,aa} + \rho_{aa,bb} + \rho_{bb,aa}, \]

\[ d = \rho_{aa,bb} - \rho_{bb,aa}, \]

\[ n = \rho_{aa,aa} - \rho_{aa,bb} - \rho_{bb,aa} + \rho_{bb,bb}. \]

Thus, \(a(\vec{r}_1, \vec{r}_2; t)\) is the single-atom excited-state population averaged over both atoms, \(d(\vec{r}_1, \vec{r}_2; t)\) is the difference in excited-state population for the two atoms, and \(n(\vec{r}_1, \vec{r}_2; t)\) is the product of the inversions of the two atoms. In new variables the equations of motion read

\[ \dot{a} = -2(\Gamma_+ + \gamma)a + \Gamma_+ - \Gamma_- d, \]

\[ \dot{d} = -2(\Gamma_+ + \gamma)d - 2\gamma(2a - 1), \]

\[ \dot{n} = -2(\Gamma_+ + \gamma)n - 2\gamma(2a - 1) + 4(\Gamma_+ + \Gamma_-) \rho_{ab,ba} + 4(\Gamma_+ - \Gamma_-) \rho_{ba}\]

\[ \dot{\rho}_{ab,ba} = - (\gamma + 2\Gamma_+ + i\Delta_{12}) \rho_{ab,ba} + (\Gamma_+ + \Gamma_-) n. \]

where we used

\[ 2\Gamma_+ = \Gamma_{11} \pm \Gamma_{22}, \]

\[ 2\Gamma_+ = \Gamma_{12} \pm \Gamma_{21}, \]

\[ \Delta_{12} = \frac{2}{\hbar}(H_{11} - H_{22}). \]

It will become clear, that the cross-coupling or correlation term \(\rho_{ab,ba}\) carries the cooperative physics.

### A. Quantum corrections in a small sample approximation

We further discuss the Eq. (15) in terms of small sample approximation, where we assume that all the coordinate dependence can be dropped from the variables describing the system, \(a\), \(n\), \(d\), and \(\rho_{ab,ab}\). That is, we neglect retardation effects of propagation of the electromagnetic field through the sample, that is, all time changes in the atomic variables propagate instantaneously through the sample. This is a good approximation as long as the “cooperative” time \(\tau_C = (\text{N}_{\text{exc}} \gamma \mu)^{-1}\), with \(\text{N}_{\text{exc}}\) the density of atoms in the excited state, \(\gamma\) the vacuum decay, and \(\mu\) a geometric factor, is longer than the maximum propagation time \(\tau_D\).

In the small sample approximation, thus, there is no difference between the atoms 1 and 2 with respect to their decay rates. That is, in Eq. (15) we set \(\Gamma_+ = \Gamma_{11} = \Gamma_{22} \equiv \Gamma\). Similarly, there is no spatial difference between the decay rates so \(\Gamma_- = \Gamma_{21} = 0\). For other decay rates we have \(\Gamma_+ = \Gamma_{12} = \Gamma_{21} \equiv \Gamma\). Finally, because in a homogeneous sample the result is invariant with respect to exchange of the indices of two atoms, we have \(\Delta_{12} = 0\). This said, the system (15) reduces to,

\[ \dot{a} = -2(\Gamma + \gamma)a + \Gamma, \]

\[ \dot{n} = -2(\Gamma + \gamma)n - 2\gamma(2a - 1) + 8\Gamma \rho_{ab,ba}, \]

\[ \dot{\rho}_{ab,ba} = - (2\Gamma + \gamma)\rho_{ab,ba} + \Gamma n. \]

Let us now discuss the decay matrices \(\Gamma_{i,j}\), where \(i, j = 1, 2\), and which we introduced in Eq. (10), in greater detail. Our discussion is based on the formalism for calculation the quantum corrections to the electric field in a responsive media, that we have previously developed in [8]. We need the exact Green’s function for the Maxwell field \(E\),

\[ D_{\mu \nu}(1,2) = \langle \langle T_{\mu}(E^\mu_-(1)E^\mu_+(2)) \rangle \rangle, \]

where \(E_0\) would be the field without medium present, as calculated along the Schwinger-Keldysh contour. Here, we abbreviate \(\vec{r}_1, t_1\) by \(\vec{1}\), etc. The Green’s function for the free field \(E_0\) is given by

\[ D_{0\mu \nu}(1,2) = \langle \langle T_{\mu}(E^\mu_-(1)E^\mu_+(2)) \rangle \rangle. \]

The contour Green’s function \(D\) contains four real-time functions, depending where on the Keldysh-Schwinger contour the two time points are located, \(D^{++}\), \(D^{-+}\), \(D^{+-}\), and \(D^{--}\), as discussed in [14]. We find approximately \(R\), \(D^{++} \approx D^{adv}\), \(D^{0+} \approx 0\), \(D^{+-} \approx D^{0-} - D^{ret}\), and \(D^{--} \approx -D^{ret}\), where \(D^{ret}\) and \(D^{adv}\) are retarded and advanced Green’s functions, respectively. A solution to the atom-field interaction can be written in terms of a Dyson equation,

\[ D^{\alpha \beta} (1,2) = - \int_{-\infty}^{+\infty} dt_1 dt_2 \int_{V_1 V_2} \frac{d^3\vec{r}_1}{d^3\vec{r}_2} D^{\alpha \beta}(1,1') P_{\mu \nu}(1',2') D^{adv}_{\nu \beta}(2',2), \]

where \(P_{\mu \nu}(1',2')\) is the propagator for the field in the vacuum, \(\bar{D}^{ret}(\vec{r}_1, \vec{r}_2; t_1, t_2)\) is the retarded Green’s function of the atom-field interaction.
where

\[ D_{a\beta}^{\text{ret}}(1, 2) = D_{a\alpha\beta}^{\text{ret}}(1, 2) - \int_{-\infty}^{+\infty} dt_1' dt_2' \]

\[ \int_{V_1 V_2} d\vec{x}_1' d\vec{x}_2' D_{a\alpha\beta}^{\text{ret}}(1, 1') P^{\text{ret}}(1', 2') D_{\beta\gamma}^{\text{adv}}(2', 2), \]

with \( D_{\beta\gamma}^{\text{adv}}(1, 2) = D_{\beta\gamma}^{\text{adv}}(2, 1) \). In Eqs. (20) and (21) we have two source (polarization) functions. In the lowest order in the atom-field coupling, the polarization function is given by a correlation function of dipole operators of noninteracting atoms,

\[ P^{(1,2)}(\vec{r}_1, \vec{r}_2; t_1, t_2) = \frac{e^{2}}{h^2} \mathcal{N}^{(1,2)} \left\{ \left\langle \sigma_1^\dagger(t_1) \sigma_j(t_2) \right\rangle \right\}, \]

and for retarded polarization,

\[ P^{(1,2)\text{ret}}(\vec{r}_1, \vec{r}_2; t_1, t_2) = \frac{e^{2}}{h^2} \mathcal{N}^{(1,2)} \Theta(t_1 - t_2) \left\{ \left\langle \sigma_1^\dagger(t_1), \sigma_j(t_2) \right\rangle \right\}. \]

In Eqs. (22) and (23) the superscript 1 stands for a one-atom source function, i.e., \( i = j \), while 2 is for a two-atom source function, and \( i \neq j \). The cumulants can be found by using the quantum-regression theorem and a Laplace transformation, as done below.

The differential equation for the single atom density-matrix cross-term, \( \rho_{ab} \), is found by tracing over the second atom,

\[ \dot{\rho}_{ab} = \dot{\rho}_{ab,aa} + \dot{\rho}_{ab,bb} = -\left( \frac{\gamma}{2} + i \Delta \right) \rho_{ab} \]

(24)

with \( \Delta = \Delta_i \equiv 2 H_{ii}/h \). Upon comparison with the same equation for a dilute gas, we find that the second-order correction \( H_{ii} \) in this case takes on the role of a detuning from resonance. Since, in general, \( H_{ii} \), along with \( \Gamma \), changes over time, a chirp of the radiated light can be expected.

We can Laplace-transform the source functions from \( \tau \) to \( \lambda \) with \( t_1 \rightarrow t + \tau \) and \( t_2 \rightarrow t \) for \( t_1 > t_2 \) (and vice versa). Thus we find for Eq. (24) (and its complex conjugate)

\[ \dot{\rho}_{ab} = \frac{\rho_{ab}(\tau = 0)}{\lambda + \gamma/2 + \Gamma + i \Delta}, \quad \dot{\rho}_{ab} = \frac{\rho_{ab}(\tau = 0)}{\lambda + \gamma/2 + \Gamma - i \Delta}, \]

(25)

Here \( \langle \sigma_i(\lambda, t) \rangle = \rho_{ab} \). Using these solutions and the quantum-regression theorem, we find for the Laplace-transformed single-atom cumulants (\( i = j \))

\[ \left\langle \sigma_1(\lambda, t) \sigma_i(t) \right\rangle = \frac{a(t)}{\lambda + \gamma/2 + i \Delta - \lambda} \left\langle \sigma_1(t) \sigma_i(\lambda, t) \right\rangle^*, \]

\[ \left\langle \sigma_i(\lambda, t) \sigma_i(t) \right\rangle = \frac{1 - a(t)}{\lambda + \gamma/2 + i \Delta + \lambda} \left\langle \sigma_i(t) \sigma_i(\lambda, t) \right\rangle^*, \]

(26)

and for the two-atom cumulants (\( i \neq j \))

\[ \left\langle \sigma_1(\lambda, t) \sigma_j(t) \right\rangle = \frac{x(t)}{\lambda + \gamma/2 + i \Delta - \lambda} \left\langle \sigma_1(t) \sigma_j(\lambda, t) \right\rangle^*, \]

\[ \left\langle \sigma_j(\lambda, t) \sigma_1(t) \right\rangle = \frac{x(t)}{\lambda + \gamma/2 + i \Delta + \lambda} \left\langle \sigma_j(t) \sigma_1(\lambda, t) \right\rangle^*, \]

(27)

In a simple case like this we can go from Laplace- to Fourier-transform by just replacing \( \lambda \) by \( i \Delta' \), where \( \Delta' \) is a frequency. Then, we find for the source functions in Fourier space

\[ P^{(1s)}(\vec{r}, \vec{r}; \Delta, t) = \frac{e^{2}}{h^2} \mathcal{N} \left( \frac{2 a(\vec{x}, t)(\gamma/2 + \Gamma)}{(\gamma/2 + \Gamma)^2 + (\Delta - \Delta')^2} \right)^2 \]

(28)

\[ P^{(2s)}(\vec{r}, \vec{r}; \Delta, t) = \frac{e^{2}}{h^2} \mathcal{N}^2 \left( \frac{2 a(\vec{x}, t)(\gamma/2 + \Gamma)}{(\gamma/2 + \Gamma)^2 + (\Delta - \Delta')^2} \right)^2 \]

(29)

\[ P^{(1r)}(\vec{r}, \vec{r}; \Delta, t) = \frac{e^{2}}{h^2} \mathcal{N} \left( \frac{1 - 2 a(\vec{x}, t)}{\gamma/2 + \Gamma + i(\Delta' - \Delta)} \right)^2 \]

(30)

where \( \vec{r}_1 - \vec{r}_2 = \vec{x} \) and \( \vec{r}_3 = \vec{r} \). Since in this case there is no external light field present, we can set \( \Delta' = 0 \).

The equations above allow us to write down closed form expressions for both \( \Gamma \) and \( \bar{\Gamma} \), where

\[ \Gamma(\Delta, \Delta) = \frac{e^{2}}{h^2} \bar{D}^{-\Delta}(\vec{r}_1 = \vec{r}_2, \Delta, t) = \]

(31a)

\[ = \frac{e^{2}}{h^2} \int_V d\vec{x} \left| \bar{D}^{-\Delta}(\vec{r}, \vec{x}, \Delta, t) \right|^2 P^{(1s)}(\vec{x}, \Delta, t) \]

\[ + \frac{e^{2}}{h^2} \int_V d\vec{x}_1 d\vec{x}_2 \bar{D}^{-\Delta}(\vec{r}, \vec{x}_1, \Delta, t) \bar{D}^{-\Delta}(\vec{r}, \vec{x}_2, \Delta, t) P^{(1s)}(\vec{x}_1, \vec{x}_2, \Delta, t) \]

\[ \bar{\Gamma}(\Delta, \Delta) = \frac{e^{2}}{h^2} \bar{D}^{-\Delta}(\vec{r}_1 \neq \vec{r}_2, \Delta, t) = \]

(31b)

\[ = \frac{e^{2}}{h^2} \int_V d\vec{x}_1 \bar{D}^{-\Delta}(\vec{r}_1, \vec{x}, \Delta, t) \bar{D}^{-\Delta}(\vec{r}_2, \vec{x}, \Delta, t) P^{(1s)}(\vec{x}, \Delta, t) + \]

\[ + \frac{e^{2}}{h^2} \int_V d\vec{x}_1 \bar{D}^{-\Delta}(\vec{r}_1, \vec{x}_1, \Delta, t) \bar{D}^{-\Delta}(\vec{r}_2, \vec{x}_2, \Delta, t) P^{(1s)}(\vec{x}_1, \vec{x}_2, \Delta, t) \]

where we use the Fourier transforms for all the functions. The \( \Delta' \) now denotes the detuning from resonance of the particular Fourier components at frequency \( \omega = \omega_{ab} + \Delta' \), where \( \omega_{ab} \) is the atomic resonance frequency. In order to solve Eqs. (31) we make an additional approximation where we neglect the coordinate dependence of the atomic variables \( a, n, \rho_{ab,ba} \) on \( \vec{r}_1, \vec{r}_2 \) in the integration. This is justified if we assume a much weaker coordinate dependence of the atomic dynamics than of the field correlations. In Ref. 3 we have discussed in great detail a way of calculating the kernel \( \bar{D}^{-\Delta}(\vec{x}_1, \vec{x}_2, \Delta, t) \). Here we just give the final result

\[ \bar{D}^{-\Delta}(\vec{x}_1, \vec{x}_2, \Delta, t) = -\frac{\hbar \omega^2}{6 \pi \epsilon_0 c^2} e^{-i q_0(\Delta) r / r}, \]

(32)

where \( r = |\vec{x}_1 - \vec{x}_2| \). Here, an average over all dipole directions is taken. The wave vector \( q_0 \) is given by

\[ q_0(\Delta) = \frac{\omega}{c} \left( 1 + i \frac{\hbar \omega}{3 \epsilon_0 c} \bar{D}^{(1r)}(r, \Delta, t) \right). \]

(33)

In Eqs. (32) and (33), \( \omega \) stands for the frequency of the emitted light.
We then obtain a self-consistent expression for the quantum corrections as follows

\[
\Gamma = \gamma \frac{a(t)}{2a(t)-1} \left( e^{\Delta/\lambda} - 1 \right) \quad (34a)
\]

\[
+ 2\gamma C^2 q^4 \frac{\Gamma_f}{\Gamma_f + \Delta^2} \rho_{ab,ba}(t) I(\bar{\zeta}(\Delta), \bar{\varrho}(\Delta)),
\]

\[
\bar{\Gamma} = 3\gamma C q \frac{\Gamma_f}{\Gamma_f + \Delta^2} a(t) I(\zeta(\Delta), \varrho(\Delta)) \quad (34b)
\]

\[
+ 2\gamma C^2 q^4 \frac{\Gamma_f}{\Gamma_f + \Delta^2} \rho_{ab,ba}(t) I(\zeta(\Delta), \varrho(\Delta)).
\]

In the expressions above both decay rates, \(\Gamma\) and \(\bar{\Gamma}\), themselves implicitly depend on the atomic variables \(a, n, d\), and \(\rho_{ab,ba}\) of Eq. (14), so they have to be self-consistently calculated. Different quantities that appear in Eq. (34) are given as follows,

\[
\Gamma_f = \frac{\zeta}{2} + \Gamma, \quad \zeta(\Delta) = \zeta_0 \frac{\Delta^2}{\Gamma^2 + \Delta^2},
\]

\[
\zeta_0 = \frac{\Gamma}{2} C q \bar{q} (2a(t) - 1), \quad \varrho(\Delta) = \varrho - \frac{\Delta}{\Gamma} \zeta(\Delta)
\]

\[
I(\zeta, \varrho) = \frac{((\zeta - 1) e^{i \varrho} \sin \varrho) + (\varrho e^{-i \varrho} \sin \varrho)^2}{\zeta + \varrho^2}.
\]

We see that the Eqs. (34) and (35) rely on two parameters: effective density, which is also known as the cooperativity parameter, \(C\), given by

\[
C = \frac{2\pi c^3 N}{\omega^3},
\]

which is proportional to the total number of atoms in a cubic wavelength of the emitted radiation, where \(N\) is the density of the atoms; and the effective radial size, \(\varrho\), of Eq. (35), proportional to the ratio of diameter \(d\) of the sample to the wave length \(\lambda\) of the emitted radiation.

Now, the role of \(\Delta = 2H_d / h\) becomes clear: As a real part of the dispersion, \(\Delta\) is related to imaginairy part \(\Gamma\), a decay rate due to cooperativity effects, via a Kramers-Kronig relationship

\[
\Delta = \frac{1}{\pi} \int_{-\infty}^{\infty} d\Delta' \frac{\Gamma(a, x, \Gamma, \Delta')}{\Delta - \Delta'}.
\]

In a two-level system, \(\Delta\) is very small, and we can set it self-consistently to zero in what follows. The detailed format of \(\Delta\) will be discussed in an upcoming publication.

### B. Cooperative Phenomena

A physical measure of the dynamics in the system is the average intensity of the emitted radiation, rather than the effective population of the excited state. The intensity of radiation is directly proportional to the term \(-\bar{a}(\tau)\) by energy conservation. One has to keep in mind that in our modeling we have neglected the time retardation effects within the sample. When detecting the emitted radiation one would have to add some delay before the radiation reaches the detectors.

We now examine short time behavior of the system when all the atoms are initially in the excited state, \(|a\rangle\). The values for the atomic variables \(a(0), x(0)\) and \(n(0)\) are then \(a(0) = 1, n(0) = 1, \rho_{ab,ba} = 0\). While the system of equations (17) provides a way of calculating the chirp \(\Delta\) from the spectral distribution \(\Gamma = \Gamma(a, n, x, \Delta)\), for simplicity we set \(\Delta = 0\) for rest of the discussed calculations. We choose as values for the effective density \(C = 10\) and the effective size \(\varrho = 10\). Different aspects of the solution of system (17), given the initial values and relevant parameters as described above, is shown in Fig. 2 (Top panel) reveals a typical signature of the cooperative enhancement to the single atom emission rate at the early stages of deexcitation. (Middle panel) depicts the quantities \(\rho_{ab,ba} = \rho_{ab,ba}(\tau)\) and \(n = n(\tau)\). Here, the most striking is the behavior of the two-atom entanglement, \(\rho_{ab,ba}\), which grows from zero to some maximum value before dropping back to zero (for times \(\ll \gamma^{-1}\), not shown here). This feature, a build-up of entanglement with a significant increase in emitted intensity per atom compared to a single atom, is a charactereristic signature of superradiance, as discussed in, e.g., Ref. [10].

One of the features of the superradiance is that the intensity per atom depends strongly on the density of an optical medium under consideration. In our theory the measure of density is the effective density \(C\), Eq. (36). We perform a series of calculations where we vary \(C\), and calculate max\{-\(\bar{a}(\tau)\)\} = \(-\bar{a}(\tau_{\text{max}})\) and \(\tau_{\text{max}}\) for each of the solutions. The results of this calculation are shown in Table I: maxima of the decay rate are proportional to the effective density \(C\), while the times at which maxima occur depend are reversely proportional to the effective density \(C\). This is in good agreement with the expected superradiance scaling, that is, the emission per atom \(-\bar{a}(\tau)\) is indeed proportional to \(C\), while the times at which the maxima are obtained scale like \(1/C\). The curves with the calculated intensity per single atom for the cooperativity parameter \(C = 10, 20, 30\) are given in Fig. 3.

| \(C\) | \(\text{max}\{-\bar{a}\}\), \(\tau_{\text{max}}\) (in \(\gamma^{-1}\)) |
|---|---|
| 10 | 57 | 0.0018 |
| 20 | 115 | 0.0008 |
| 30 | 172 | 0.0006 |

**TABLE I**: Dependence of the maximum of the intensity per atom \(-\bar{a}\) and the scaled time \(\tau_{\text{max}}\), at which the maximum occurred, as a function of \(C\). The quantity \(-\bar{a}\) is a linear function of \(C\), and thus confirming the superradiant nature of the decay. The times \(\tau_{\text{max}}\) at which the maximum of the radiation is achieved, scale like \(1/C\).
subradiant, $\rho_{--}$, for the population of anti-symmetric two-atom state, and a superradiant, $\rho_{++}$, for the symmetric two-atom state, where $|\pm\rangle = 1/\sqrt{2}(|ab\rangle \pm |ba\rangle)$. This is permissible if the size of the atomic sample is small, because the effects of change of phase due to propagation can be neglected. In terms of atomic variables $a(\tau), n(\tau)$ and $\rho_{ab,ba}(\tau)$, these matrix elements are $\rho_{\pm\pm} = 1/4(1-n(\tau)\pm 4\rho_{ab,ba}(\tau))$. The behavior of the matrix elements under the conditions exhibiting superradiant behavior, Eq. (17), and with the cooperativity parameter $C = 10$ is shown in Fig. 4. Here we observe that the dipole-dipole interaction, which is built into our model, indeed contributes to populating the anti-symmetric two-atom state, whereas the population of $|+\rangle$ and $|-\rangle$ equalizes for long times when decoherence overrules correlation. This feature is absent in standard treatments of superradiation, e.g. [16].

FIG. 4: Superradiant, $\rho_{++}$, and subradiant $\rho_{--}$ density matrix element in logarithmic time for a dense optical media with $C = 10$. The solution of system (17) shows that in a superradiant decay both, superradiant and subradiant, states get populated. As expected, there is a significant difference between the two, as long as there is an entanglement present, $\rho_{ab,ba}(t) \neq 0$.

IV. CORRELATION AND ENTANGLEMENT

In this paragraph we would like to clarify the respective roles of correlation and entanglement in superradiance. Correlation, i.e., a two-atom cooperative effect, is stored in the non-diagonal two-atom matrix element, $\rho_{ab,ba}$, and can be measured as the energy splitting between the $|+\rangle$ and $|-\rangle$ state. This quantity clearly is responsible for the build-up of superradiation (see, e.g., Eqs. (15), (17), and (34)). Entanglement, however, defined to be a quantity describing how far the system is away from a product state $\rho_{\text{prod}} = \sum_i \rho_i^{(1)} \otimes \rho_i^{(2)}$, where $\rho_i^{(1/2)}$ are the single atom density matrices. Wootters [17] has found a measure for the entanglement of two systems. It turns out that in all two-atom superradiant systems, even those with a maximum grade of coherence, there is

FIG. 3: Superradiant intensity per atom, $-\dot{a}(\tau)$, in dense optical media of cooperativity parameter $C = 10, 20, 30$. Greater the cooperativity parameter, greater the maximum momentary deexcitation rate $\Gamma = \Gamma(\tau)$, see also Table 4.
no entanglement present. The proof is easy, since \( \rho_{abba} \) always has to be smaller or equal to \( \sqrt{\rho_{aa}\rho_{bb}} \), where \( \rho_{aa} \) and \( \rho_{bb} \) are the single-atom populations.

Obviously, at this time it is not possible to make any final statement about the entanglement of more than two atoms with cooperative interactions, but since we treat only effects up to second order in the atomic interaction exactly, it is quite clear that also in our system there is no entanglement present. In conclusion, superradiance is an effect based on correlation, but not on entanglement.

V. CONCLUSION

We have presented a method for modeling the cooperative effects in a dense optical medium, based on an effective two-atom two-level model. In its simplest version, a small sample approximation, we are able to obtain a system of equations which describes the evolution of a two-atom two-level density matrix. Solving the system for an initially inverted system, we are able to predict the cooperative behavior with all the features of superradiance: coherent build-up of entanglement, with the decay rate up to two orders of magnitude greater than the spontaneous decay rate. Additionally, the maximum of the decay rate \( \Gamma \) scales linearly with the number of particles indicating that the maximum intensity of the deexcitation pulse scales as \( N^2 \), the square of the number of particles. Our model requires two parameters, the cooperativity parameter \( C \), and the effective size of the system in terms of the emitted wavelength. Theory predicts two novel phenomena: chirp, or the change in transition frequency due to the cooperative phenomena and subradiance, both of which are outside a standard description of superradiance. Accounting for the shape of the optical medium in the formalism, and in particular analysis of cigar-shaped atomic sample, will be subject of future work.

[1] R. Dicke, Phys. Rev. 93, 99 (1953).
[2] N. E. Rehler and J. H. Eberly, Phys. Rev. A 3, 1735 (1971).
[3] M. Fleischhauer, S. F. Yelin, and M. D. Lukin, Opt. Comm. 179, 395 (2000).
[4] M. Fleischhauer and M. D. Lukin, Phys. Rev. Lett. 65, 5094 (2000).
[5] M. Fleischhauer and M. D. Lukin, Phys. Rev. A 65, 022314 (2002).
[6] R. Walsworth, S. F. Yelin, and M. D. Lukin, Opt. Phot. News. 13, 50 (2002).
[7] M. G. Moore and P. Meystre, Phys. Rev. Lett. 83, 5202 (1999).
[8] M. Fleischhauer and S. Yelin, Phys. Rev. A 59, 2427 (1999).
[9] L. Keldysh, Zh. Eksp. Teor. Fiz 47, 1515 (1964), English translation in JETP Lett. 20, 2018 (1985).
[10] C. Gardiner, Handbook of Statistical Methods (Springer-Verlag, Berlin, 1985).
[11] M. Fleischhauer, Phys. Rev. A 50, 2773 (1994).
[12] P. Schwendimann, Z. Phys. 265, 267 (1973).
[13] R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302 (1971).
[14] A. Fetter and J. Walecka, Quantum Theory of Many-particle Systems (McGraw-Hill, New York, 1971).
[15] P. Meystre and M. Sargent, Elements of Quantum Optics (Springer, New York, 1999).
[16] M. Gross and S. Haroche, Phys. Rep. 93, 302 (1982).
[17] W. K. Wootters, Phys. Rev. Lett. 80, 2245 (1998).
[18] \( \tau_C \) gives a good estimate for the shortest possible superradiance timescales. The actual superradiance time \( \tau_{sr} \), i.e., the time for the buildup of radiation, is the one that finally has to be short compared to the propagation time.

Usually, \( \tau_{sr} > \tau_C \).