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Superfluorescence and cooperative frequency shift

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We discuss from first principles the cooperative decay of a system of two-level atoms, initially prepared in an uncorrelated excited state with population inversion N, and we give the conditions under which the superfluorescence effect occurs. Describing the atomic system in terms of collective variables, we derive a master equation for the reduced atomic density operator, which gives rise both to a damping and to a time-dependent frequency shift in the dynamics of collective modes. The coupled equations of motion are solved with a self-consistent approach. It is found that the system goes through a nonexponential decay if the maximum length of the active volume is smaller than a "cooperation range" and larger than a "threshold length," in agreement with the one-mode theory. The radiation burst has a time width proportional to N^{-1}, and its intensity is proportional to N^2. Specializing to a pencil-shaped volume, we find that only two atomic modes need to be considered; in this case, the average emitted radiation is all condensed in the two diffraction patterns of the opposite axial modes.

I. INTRODUCTION

The first description of the cooperative phenomena which occur when N excited atoms radiate was made by Dicke,[1] who showed that a radiated intensity proportional to N^2 takes place under the following assumptions: (a) The atoms are confined to a volume much smaller than one wavelength; (b) the two-level atoms of the system are coherently prepared in an initial correlated state with macroscopic polarization (superradiance). The first assumption was later removed by Eberly and Rehler,[2] who generalized Dicke's description to a large system.

We shall call "superfluorescence" the effect which takes place, under certain conditions, starting from an uncorrelated state (incoherent pumping). While the radiation process in superradiance is essentially classical, cooperative spontaneous emission (CSE), in superfluorescence it is intrinsically a quantum process. In fact, the system starts radiating by ordinary fluorescence, and then eventually evolves spontaneously toward a correlated state, in which it radiates proportionally to N^2.

Superfluorescence has been treated by Bonifacio, Schwendimann, and Haake, assuming strong coupling with only one e.m. mode, "the Dicke end fire mode."[3,4] Emission of radiation into other modes, and escape of radiation from the active volume V_C, where atoms are enclosed, were taken into account phenomenologically, inserting relaxation terms into the equations of motion for the density operator.

In this paper[5] we report a derivation from first principles of a many-mode theory of cooperative decay and superfluorescence effect. The central ideas are as follows:

(i) The e.m. field is quantized in a volume V, much greater than V_C. Irreversibility is obtained by letting $V \to \infty$ at the end of calculations.

(ii) The dynamics of the atomic system are studied through the collective atomic modes of V_C.

In this way, we are able to arrive at a m.e. (master equation) for the reduced density operator of the atomic variables. We evaluate the coefficients of the m.e. in the Markoffian approximation and for a simple geometry of V_C. We give the equation for the emitted radiation and the conditions under which superfluorescence occurs.

Furthermore, we find that the frequency of the collective modes presents a time-dependent cooperative shift (frequency chirping) proportional to N, which is added to the single-atom Lamb shift. This frequency chirping has been described up to now in the framework of semiclassical theory for a single atom[6] or for N atoms confined to a small volume[7] ($V_C \ll \lambda^3$). We show that in a quantum theory chirping does not arise for a single atom,[8] while it is present for many atoms and also for $V_C \gg \lambda^3$ whenever certain conditions about the excitation and the geometry of V_C are met.

Finally, we specialize to a pencil-shaped volume. We then find that, if the conditions of Ref. 3 are satisfied, superfluorescence takes place into the diffraction patterns of the "end fire modes" and the results of Ref. 3 are correct.

II. PHYSICAL MODEL

We assume that the system is constituted of N two-level atoms, interacting among themselves and with the external world only through the e.m. field. Unless otherwise specified, we disregard any interaction via phonons, collisions, etc. It is convenient, for mathematical reasons, to assume that the atoms are placed in a regular cubic lattice, one at each vertex, with spacing d. We shall see that this assumption does not imply any physical restrictive condition, as there are many atoms in a wavelength. We denote with L_i, $i=x,y,z$, the dimensions of the parallelepiped occupied by the atoms, and with V_C its volume: $V_C = Nd^3 = \prod_i L_i$.

Let Ω be the frequency difference between atomic levels, and λ and k_0 the corresponding radiation wavelength and wave vector *in vacuo*, respectively. We quantize the e.m. field in a box of volume V, $V \gg V_C$. The quantization is made on the free field in the Coulomb gauge, and the modes $\vec{k}$ are transverse. We neglect the electrostatic dipole interaction. In the dipole approximation we have the Hamiltonian

$$H = H_A + H_F + H_{AF}$$
$$= \hbar\Omega \sum_i r_{3,i} + \hbar \sum_{\vec{k}} \omega_{\vec{k}} a_{\vec{k}}^\dagger a_{\vec{k}} + \hbar V^{-1/2} \sum_{\vec{k},i} g_{\vec{k}} \{[a_{\vec{k}} r_i^+ \exp(i\vec{k}\cdot\vec{x}_i) + \text{H.c.}] + [a_{\vec{k}}^\dagger r_i^+ \exp(-i\vec{k}\cdot\vec{x}_i) + \text{H.c.}]\}, \quad (1)$$

where $a_{\vec{k}}^\dagger$ and $a_{\vec{k}}$ are the creation and annihilation operators of the field mode $\vec{k}$, $[a_{\vec{k}}^\dagger, a_{\vec{k}'}] = \delta_{\vec{k},\vec{k}'}$; $r_i^\pm$ and $r_{3,i}$ are the spin operators referring to atom i,

$$[r_{3,i}, r_j^\pm] = \pm r_i^\pm \delta_{i,j}, \quad [r_i^+, r_j^-] = 2r_{3,i}\delta_{i,j};$$

$\vec{x}_i$ is the space position of atom i; $g_{\vec{k}}$ is the coupling constant between the field mode $\vec{k}$ and one atom, $g_{\vec{k}} = \Omega(2\hbar\omega_{\vec{k}})^{-1/2}[\mu_{12}^2 - (\hat{k}\cdot\vec{\mu}_{12})^2]^{1/2}$, with $\vec{\mu}_{12}$ the dipole matrix element.

A. Collective operators

We could now describe the dynamics of the system in terms of single-atom operators, as it is expressed in Hamiltonian (1). However, with this approach, we have complicated coupled equations[9] in the single-dipole operators from which we cannot clearly see the "phasing" of the atoms due to field emission on a wavelength λ. These difficulties are partially overcome if one goes to collective dipole operators.[10] We define

$$R_{\vec{\alpha}}^\pm = \sum_{i=1}^N r_i^\pm \exp(\pm i\vec{\alpha}\cdot\vec{x}_i),$$
$$R_{3,\vec{\alpha}} = \sum_{i=1}^N r_{3,i} \exp(-i\vec{\alpha}\cdot\vec{x}_i), \quad (2a)$$

where $\vec{\alpha}$ labels the modes of volume V_C, and each component α_i verifies $\alpha_i = 2\pi L_i^{-1} n_i$, n_i integer, $-\pi d^{-1} < \alpha_i \leq \pi d^{-1}$, $i=x,y,z$.

Obviously, the number of modes $\vec{\alpha}$ is N; so Eqs. (2a) are simply a linear transformation from the N operators r_i to the N operators $R_{\vec{\alpha}}$. Using the fact that

$$N^{-1}\sum_i \exp[i(\vec{\alpha}-\vec{\alpha}')\cdot\vec{x}_i] = \delta_{\vec{\alpha},\vec{\alpha}'},$$
$$N^{-1}\sum_{\vec{\alpha}} \exp[i\vec{\alpha}\cdot(\vec{x}_i - \vec{x}_j)] = \delta_{i,j}, \quad (2b)$$

transformation (2a) can be easily inverted to express single-atom operators in terms of the collective ones, i.e.,

$$r_{3,i} = N^{-1}\sum_{\vec{\alpha}} R_{3,\vec{\alpha}} \exp(+i\vec{\alpha}\cdot\vec{x}_i),$$
$$r_i^\pm = N^{-1}\sum_{\vec{\alpha}} R_{\vec{\alpha}}^\pm \exp(\mp i\vec{\alpha}\cdot\vec{x}_i). \quad (3)$$

The commutation relations for $R_{\vec{\alpha}}$ are easily obtained from the definition (2a) and the commutators for the r_i as follows:

$$[R_{\vec{\alpha}}^+, R_{\vec{\alpha}'}^-] = 2R_{3,\vec{\alpha}-\vec{\alpha}'}, \quad [R_{3,\vec{\alpha}'}, R_{\vec{\alpha}}^\pm] = \pm R_{\vec{\alpha}\mp\vec{\alpha}'}^\pm,$$
$$[R_{\vec{\alpha}}^+, R_{\vec{\alpha}'}^+] = [R_{\vec{\alpha}}^-, R_{\vec{\alpha}'}^-] = [R_{3,\vec{\alpha}}, R_{3,\vec{\alpha}'}] = 0. \quad (4)$$

Hamiltonian (1) can now be expressed in terms of the collective operators obtaining

$$H = H_F + H_A + H_{AF}$$
$$= \hbar \sum_{\vec{k}} \omega_{\vec{k}} a_{\vec{k}}^\dagger a_{\vec{k}} + \hbar\Omega R_3 + \hbar V^{-1/2} \sum_{\vec{k},\vec{\alpha}} g_{\vec{k}} \{[a_{\vec{k}} R_{\vec{\alpha}}^+ f(\vec{k}-\vec{\alpha}) + \text{H.c.}] + [a_{\vec{k}}^\dagger R_{\vec{\alpha}}^+ f^*(\vec{k}+\vec{\alpha}) + \text{H.c.}]\}, \quad (5)$$

where R_3 stays for $R_{3,0}$, and $f(\vec{k}-\vec{\alpha})$ is given by

$$f(\vec{k}-\vec{\alpha}) = N^{-1}\sum_i \exp[i(\vec{k}-\vec{\alpha})\cdot\vec{x}_i]. \tag{6a}$$

Since the atoms are placed at the vertices of a cubic lattice with spacing d, if we place the coordinate origin in the center of V_C, $f(\vec{k}-\vec{\alpha})$ becomes a real function, i.e.,

$$f(\vec{k}-\vec{\alpha}) = N^{-1}\prod_{i=1}^{3}\frac{\sin[(\vec{k}-\vec{\alpha})_i L_i/2]}{\sin[(\vec{k}-\vec{\alpha})_i d/2]}. \tag{6b}$$

Inspection of Eq. (6b) will show that the function f is nothing but the diffraction pattern of a cubic grating. For a fixed $\vec{\alpha}$, $f(\vec{k}-\vec{\alpha})$ reaches its maxima for $\vec{k}=\vec{\alpha}+\vec{g}$, where $\vec{g}$ is a vector of the reciprocal lattice: $g_i = 2\pi d^{-1}m_i$, m_i integer. The $\vec{k}$ range around the maxima (where $f=1$) is given by $\Delta k_i \sim 2\pi L_i^{-1}$. One can see that whenever $L_i \gg d$, i.e., when $N\gg 1$, the maxima are very well separated one from another and one can replace Eq. (6b) with

$$f(\vec{k}-\vec{\alpha}) = \sum_{\vec{g}}\prod_{i=1}^{3}\operatorname{sinc}\{[\vec{k}-(\vec{\alpha}+\vec{g})]_i L_i/2\}, \tag{6c}$$

where $\operatorname{sinc}x = x^{-1}\sin x$.

In the optical domain, more precisely when $\lambda \gg d$, we shall consider for the sake of simplicity only the $\vec{g}=0$ term in (6c), i.e.,

$$f(\vec{k}-\vec{\alpha}) = \prod_{i=1}^{3}\operatorname{sinc}[(\vec{k}-\vec{\alpha})_i L_i/2]. \tag{6d}$$

Indeed, it can be easily seen that the $\vec{g}\neq 0$ terms of (6c) give to the $a_{\vec{k}}^{\dagger} R_{\vec{\alpha}}^{\pm} f(\vec{k}\mp\vec{\alpha})$ terms of the interaction Hamiltonian a time behavior characterized by frequencies larger than cd^{-1}, so the $\vec{g}\neq 0$ terms can be disregarded for $d \ll \lambda$, i.e., when one has many atoms in a wavelength. In this limit the results we obtain are independent of the lattice assumption of our model.

III. MASTER EQUATION

The motion of the total system, atoms and field, is governed by the Liouville equation for the density operator W:

$$\dot{W} = i\mathfrak{L}W, \quad \mathfrak{L} = \hbar^{-1}[H, \ ]. \tag{7}$$

If one needs to evaluate the mean values of quantities referring to only one subsystem (i.e., atoms or field), he can trace away from W everything not referring to the particular subsystem, obtaining in this way a reduced density operator. We shall use the reduced density operators of the atoms and of the field, denoted, respectively, by W_A and W_F, i.e.,

$$W_A = \mathrm{tr}_F W, \quad W_F = \mathrm{tr}_A W. \tag{8}$$

Starting from the initial condition $W(0)$, it is possible, through the projector technique introduced by Zwanzig,[11] to obtain a closed non-Markoffian equation for W_A or W_F. Once one of the reduced density operators is known, it is easy to derive the other with some calculation. In our case it is suitable to write the m.e. in terms of W_A, i.e., to trace out the field in deriving the atomic dynamics and to calculate the field properties later.

For the initial conditions we assume that $W(0)$ can be factorized,

$$W(0) = W_A(0)\otimes W_F(0), \tag{9}$$

and that the field is in the vacuum state, so that

$$W_F(0) = |0\rangle\langle 0|. \tag{10}$$

With the initial conditions (9) and (10) we calculate in Appendix A the evolution of W_A. To arrive at the m.e. (12), we have to apply the Born approximation, i.e., a second-order perturbation in the interaction Hamiltonian which amounts to disregarding the reaction of the field back on the atoms.

A discussion of the Born approximation is given in Ref. 3, and the considerations made there are fully valid in our case as well. The limit of validity of the Born approximation can be shown to be

$$L_{\max} \ll l_c = (cV_C/4\pi N\lambda^2\gamma_0)^{1/2}, \tag{11}$$

with l_c the cooperative length of the atomic system[12] and γ_0^{-1} the single-atom fluorescence decay time.

In the interaction representation given by the unitary operator $U(t) = \exp[-i\hbar^{-1}(H_A+H_F)t]$, the evolution of $\tilde{W}_A$, $\tilde{W}_A = UW_AU^{\dagger}$, is given by

$$\dot{\tilde{W}}_A(t) = \sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t d\tau\{A_{\vec{\alpha},\vec{\alpha}'}^{-}(\tau)[\tilde{R}_{\vec{\alpha}}^{-}, \tilde{W}_A(t-\tau)\tilde{R}_{\vec{\alpha}'}^{+}] + \mathrm{H.c.}\} + \sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t d\tau\{A_{\vec{\alpha},\vec{\alpha}'}^{+}(\tau)[\tilde{R}_{\vec{\alpha}}^{+}, \tilde{W}_A(t-\tau)\tilde{R}_{\vec{\alpha}'}^{-}] + \mathrm{H.c.}\}, \tag{12}$$

where

$$A_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau) = (2\pi)^{-3}\int d^3\vec{k}\, g_{\vec{k}}^2 f(\vec{k}\pm\vec{\alpha})f(\vec{k}\pm\vec{\alpha}')\exp(\pm i\eta_{\vec{k}}^{\pm}\tau), \tag{13}$$

with $\eta_{\vec{k}}^{\pm} = \Omega \pm \omega_{\vec{k}}$.

Equation (12) is the non-Markoffian m.e. governing the dynamics of the atoms. The time lag to be considered in integration (12), i.e., the range of the memory of the system, depends on $A_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau)$.

In general, we can say that $A_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau)$ is different from 0 in a time interval $\Delta\tau_{\vec{\alpha},\vec{\alpha}'}$ such that

$$\Delta\tau_{\max} \sim c^{-1}L_{\max}; \tag{14}$$

i.e., the maximum time interval to take into ac-

count in Eq. (11) is of the order of the maximum transit time of radiation in V_C. This can be seen by substituting Eq. (6d) into Eq. (13) and going to polar coordinates in $\vec{k}$ space. We have for $A^{\pm}_{\vec{\alpha},\vec{\alpha}'}$ (and similarly for A^{+}) such integrals as

$$\prod_{i=1}^{3}\int d\Omega_{\vec{k}}^{\pm}\int g_{\vec{k}}^{\pm 2}\,k^2\,\mathrm{sinc}[(k\hat{k}-\vec{\alpha})_i\,L_i/2]$$

$$\times\,\mathrm{sinc}[(k\hat{k}-\vec{\alpha}')_i L_i/2]\exp(i\eta_{\vec{k}}^{\pm}\tau)\,dk\,,$$

where $d\Omega_{\vec{k}}^{\pm}$ is the differential solid angle around the unit vector $\hat{k}$. Integration over k modulus, for a fixed $\hat{k}$, is nothing but a Fourier transform that can be done by making the convolution in time domain of the transforms of the sinc functions. Since

$$\int \mathrm{sinc}[(k\hat{k}-\vec{\alpha})_i L_i/2]\exp(ick\tau)\,dk = 0$$

for $|\tau|>(2c)^{-1}\hat{k}_i L_i$, the convolution will be zero for $\tau > c^{-1}\sum_i \hat{k}_i L_i$, i.e., for $\tau > c^{-1}L_{\max}$. Because this property is not changed by the integration in $d\Omega_{\vec{k}}^{\pm}$, we can make the statement (13).

Furthermore, we can explicitly evaluate $A^{\pm}_{\vec{\alpha},\vec{\alpha}'}(\tau)$ (or A^{+}), with $\vec{\alpha}=\vec{\alpha}'$, whenever we can assume it is possible to replace $|\vec{k}|$ in Eq. (13) with

$$|\vec{k}| \simeq \vec{k}\cdot\hat{\alpha}\,. \tag{15}$$

The choice of this approximation stems from the fact that $f^2(\vec{k}-\vec{\alpha})$ is strongly peaked at $\vec{k}=\vec{\alpha}$. In Appendix B we give the discussion of the conditions of validity of Eq. (15), and all the calculations. We state here only the result

$$A^{\pm}_{\vec{\alpha},\vec{\alpha}}(\tau) \simeq V_C^{-1}\,g_{\vec{\alpha}}^{\pm 2}\exp[(\pm i\eta_{\vec{\alpha}}^{\pm}-\chi_{\vec{\alpha}}^{\pm})\tau]\,, \tag{16a}$$

where

$$\chi_{\vec{\alpha}}^{\pm} = c\sum_{i=1}^{3}\hat{\alpha}_i L_i^{-1}. \tag{16b}$$

We see that the damping is given by the inverse of the transit time of radiation in V_C along the direction of the mode considered. Let us now make some remarks on $A^{\pm}_{\vec{\alpha},\vec{\alpha}'}(\tau)$ with $\vec{\alpha}\neq\vec{\alpha}'$. These coefficients arise from the fact that in V_C there are no pure e.m. modes $\vec{\alpha}$, but rather quasimodes $\vec{\alpha}$. Various orders of diffraction of the e.m. quasimode $\vec{\alpha}$ interact with the atomic modes $\vec{\alpha}'$, giving rise to a coupling among the atomic modes themselves when one traces away the field variables. We stress the fact that this kind of mode coupling, represented by the terms with $\vec{\alpha}\neq\vec{\alpha}'$ in m.e. (12), does not depend on the nonlinearity of the phenomena associated with a spin system; in fact, one can see that it is also present for a system of harmonic oscillators (which can, in principle, be exactly solved with a diagonalization). However, as we discuss in Appendix B, the contribution of the terms with $\vec{\alpha}\neq\vec{\alpha}'$ to the atomic decay is very small with respect to the ones with $\vec{\alpha}=\vec{\alpha}'$, and it goes to zero in the Markoffian limit. Furthermore, since the role of the $\vec{\alpha}\neq\vec{\alpha}'$ terms can be neglected also for what concerns the unitary part of the atomic evolution (see Appendix B and the discussion on the spin pseudo-Hamiltonian), we are led to disregard in the following the terms with $\vec{\alpha}\neq\vec{\alpha}'$ in our master equation.

A. Markoffian master equation

If we assume that the atomic system has a dynamics with a characteristic time much larger than $\tau_{\max}$ of Eq. (14), i.e., $\tilde{W}_A$ is approximately constant during the time τ in which the quantities $A^{\pm}_{\vec{\alpha},\vec{\alpha}}$ are different from zero, we can take the commutators evaluated at $\tau=0$ out of the time integral of m.e. (12). We have

$$\dot{W}_A(t) = -i\Omega[R_3, W_A(t)] + \tfrac{1}{2}\sum_{\vec{\alpha}}\{\Gamma_{\vec{\alpha}}^{-}[R_{\vec{\alpha}}^{-}, W_A(t)R_{\vec{\alpha}}^{+}] + \mathrm{H.c.}\}$$

$$+\tfrac{1}{2}\sum_{\vec{\alpha}}\{\Gamma_{\vec{\alpha}}^{+}[R_{\vec{\alpha}}^{+}, W_A(t)R_{\vec{\alpha}}^{-}] + \mathrm{H.c.}\}\,, \tag{17}$$

where we have gone back from $\tilde{W}_A$ of the interaction representation to W_A. The coefficients $\Gamma_{\vec{\alpha}}^{\pm}$ of Eq. (17) are given by

$$\Gamma_{\vec{\alpha}}^{\pm} = 2\int_0^{\infty} A^{\pm}_{\vec{\alpha},\vec{\alpha}}(\tau)\,d\tau$$

$$= (2\pi)^{-3}\int g_{\vec{k}}^{\pm 2}\,\delta(c|\vec{k}|\pm\Omega)\,f^2(\vec{k}-\vec{\alpha})\,d^3\vec{k}$$

$$\pm i\,(2\pi)^{-3}\,\mathrm{P.}\int g_{\vec{k}}^{\pm 2}\,\frac{2}{\eta_{\vec{k}}^{\pm}}\,f^2(\vec{k}-\vec{\alpha})\,d^3\vec{k}, \tag{18}$$

where P. means to take the principal part. Let us write $\Gamma_{\vec{\alpha}}^{\pm}$ as

$$\Gamma_{\vec{\alpha}}^{\pm} = \gamma_{\vec{\alpha}}^{\pm} + i\Omega_{\vec{\alpha}}^{\pm}\,, \tag{19}$$

where we have defined $\gamma_{\vec{\alpha}}^{\pm}$ and $\Omega_{\vec{\alpha}}^{\pm}$, respectively, as the real and imaginary parts of $\Gamma_{\vec{\alpha}}^{\pm}$. Note that $\gamma_{\vec{\alpha}}^{+}=0$. The evaluation of $\gamma_{\vec{\alpha}}^{-}$ and $\Omega_{\vec{\alpha}}^{\pm}$ can be done substituting Eqs. (16) in Eq. (18) and performing the time integral, or it can be evaluated directly from Eq. (18). This second way seems to us easier for evaluating $\gamma_{\vec{\alpha}}^{-}$ in a pencil-shaped geometry.

Let us now make some substitutions which will demonstrate the cooperative effect in m.e. (17). We define a cooperative polarization $S_{\vec{\alpha}}^{-}$ as

$$S_{\vec{\alpha}}^{-} = \sum_{i\neq j} r_i^{+} r_j^{-}\exp[i\vec{\alpha}\cdot(\vec{x}_i-\vec{x}_j)]\,. \tag{20a}$$

Remembering that $r_i^{+}r_i^{-} = r_{3} + \frac{1}{2}$, one can verify from Eq. (20a) and definition (2) that

$$S_{\vec{\alpha}}^{-} = R_{\vec{\alpha}}^{+}R_{\vec{\alpha}}^{-} - (\tfrac{1}{2}N + R_3). \tag{20b}$$

$S_{\vec{\alpha}}^{-}$ represents the degree of "phasing" between the

polarization of different atoms. For a completely random situation, typical of a group of uncorrelated atoms, one has $\langle S_{\vec{\alpha}}^{\pm}\rangle = 0$ for all $\vec{\alpha}$.

As we shall see later, the superfluorescence effect is linked to the buildup of such correlation; i.e., $\langle S_{\vec{\alpha}}^{\pm}\rangle$ for some resonating $\vec{\alpha}$ becomes different from 0, even if they were so at the initial time.

Substituting Eqs. (19) and (20b) into m.e. (17), we have

$$\dot{W}_A = -i(\Omega+\Omega')[R_3, W_A] + i\tfrac{1}{2}\sum_{\vec{\alpha}} \Omega_{\vec{\alpha}}^{\pm}[S_{\vec{\alpha}}^{\pm}, W_A] + \tfrac{1}{2}\sum_{\vec{\alpha}} \gamma_{\vec{\alpha}}^{\mp}\{[R_{\vec{\alpha}}^{\mp}, W_A R_{\vec{\alpha}}^{\pm}] + \text{H.c.}\}, \tag{21}$$

where we have set

$$\Omega' = \tfrac{1}{2}\sum_{\vec{\alpha}} (\Omega_{\vec{\alpha}}^{+} - \Omega_{\vec{\alpha}}^{-}), \tag{22}$$

$$\Omega_{\vec{\alpha}}^{\pm} = \Omega_{\vec{\alpha}}^{+} + \Omega_{\vec{\alpha}}^{-}. \tag{23}$$

Equation (21) represents the generalization of the one in Ref. (3) to a many-mode case, and it is the starting point for solving the dynamics of C.S.E., whenever the Markoffian approximation holds.

Let us now consider the meaning of the different terms in Eq. (21) and the value of their coefficients. The quantity Ω' in the first term of Eq. (21) gives a constant frequency shift to the naked frequency Ω, giving rise to the usual frequency renormalization of a single atom. In fact, substitution of Eq. (18) in Eq. (23) gives

$$\Omega' = (2\pi)^{-3}\,\mathrm{P.}\int g_{\vec{k}}^2 \left(\frac{1}{\Omega-\omega_{\vec{k}}} + \frac{1}{\Omega+\omega_{\vec{k}}}\right) d^3\vec{k}, \tag{22'}$$

where we have made use of the fact that $\sum_{\vec{\alpha}} f^2(\vec{k}-\vec{\alpha}) = 1$, as can be proved from definition (6a). Ω' given by (22′) is simply the Bethe part of the Lamb shift of a two-level atom.

A completely different role is played by the second term; in fact, the frequency shift caused by $\Omega_{\vec{\alpha}}^{\pm}$ comes from cooperative effects, as can be seen in the extreme situation of only one atom present, when $S_{\vec{\alpha}}^{\pm}$ disappears. Under condition (15), $\Omega_{\vec{\alpha}}^{\pm}$ can be calculated immediately from Eqs. (16) and (18); we have

$$\Omega_{\vec{\alpha}}^{\pm} = 2g_{\vec{\alpha}}^2 V_C^{-1}\left(\frac{\eta_{\vec{\alpha}}^{-}}{(\eta_{\vec{\alpha}}^{-})^2+\chi_{\vec{\alpha}}^2} - \frac{\eta_{\vec{\alpha}}^{+}}{(\eta_{\vec{\alpha}}^{+})^2+\chi_{\vec{\alpha}}^2}\right). \tag{23'}$$

In Eq. (23′) we have omitted the $\vec{\alpha}$-independent constant, for the calculation of which we have to put a cutoff in the frequency.

From Eq. (23′) we see that $\Omega_{\vec{\alpha}}^{\pm}$ goes to zero for resonant modes, and reaches the maximum positive and the minimum negative values for the modes just below and just above resonance.

To get an insight into the effects of this term let us calculate the dynamics of $R_{\vec{\alpha}}^{\pm}$ with m.e. (21). We have

$$\langle \dot{R}_{\vec{\alpha}}^{\mp}\rangle = -i\Omega_t\langle R_{\vec{\alpha}}^{\mp}\rangle - i\sum_{\vec{\alpha}'} \Omega_{\vec{\alpha}'}^{\pm}\langle R_{3,\vec{\alpha}'-\vec{\alpha}} R_{\vec{\alpha}'}^{\mp}\rangle + (\text{damping terms}), \tag{24}$$

where we have confined in Ω_t the $\vec{\alpha}$-independent coefficients. An analogous expression holds for $\langle \dot{R}_{\vec{\alpha}}^{\pm}\rangle$. The second term on the right-hand side of Eq. (24) in the self-consistent-field approximation, becomes

$$-i\sum_{\vec{\alpha}'} \Omega_{\vec{\alpha}'}^{\pm}\langle R_{3,\vec{\alpha}'-\vec{\alpha}}\rangle\langle R_{\vec{\alpha}'}^{\mp}\rangle \simeq -i\Omega_{\vec{\alpha}}^{\pm}\langle R_3\rangle\langle R_{\vec{\alpha}}^{\mp}\rangle, \tag{25}$$

where we have disregarded $\langle R_{3,\vec{\alpha}'-\vec{\alpha}}\rangle$ with $\vec{\alpha}\neq\vec{\alpha}'$, which amounts to the assumption that all $\langle r_{3,j}\rangle$ are equal. We can then define an instantaneous frequency shift given by $\Omega_{\vec{\alpha}}^{\pm}\langle R_3\rangle$. From Eqs. (24) and (25), one can see that the $\Omega_{\vec{\alpha}}^{\pm}$ term in Eq. (17) gives rise to a time-dependent frequency shift (chirping) when $\langle R_3\rangle$ goes from the initial value $\frac{1}{2}\overline{N}$ [for $\frac{1}{2}(\overline{N}+N)$ excited atoms] to $-\frac{1}{2}N$. The total frequency sweep during emission is of the order of $\Omega_{\vec{\alpha}}^{\pm}N$.

The last term in m.e. (17) gives a nonunitary time evolution. In the equation of motion, as will be shown below, it gives rise to the decay of excitation and is comprehensive of both the Wigner-Weisskopf exponential decay and the cooperative superradiant decay.

From Eqs. (16a), (18), and (19), we have

$$\gamma_{\vec{\alpha}}^{\mp} = g_{\vec{\alpha}}^2 V_C^{-1}\chi_{\vec{\alpha}}^{\pm}/[(\eta_{\vec{\alpha}}^{\mp})^2+\chi_{\vec{\alpha}}^2]. \tag{26}$$

One can verify from definition (19), or from the particular form (26), that

$$\sum_{\vec{\alpha}} \gamma_{\vec{\alpha}}^{\mp} = \pi^{-1}\langle g_{k_0}^2\rangle \frac{\Omega^2}{c^3} = \gamma_0, \tag{27}$$

where γ_0 is the Wigner-Weisskopf decay constant, and $\langle g_{k_0}^2\rangle$ is the average over all directions of the coupling constant evaluated at resonance.

Let us note that the results obtained for a uniform dipole orientation are valid also for a randomly distributed dipole orientation, provided we make the replacement $g_{\vec{k}} = g_k = \langle g_k^2\rangle^{1/2}$, where $k = |\vec{k}|$. For resonant modes, Eq. (26) gives $\gamma_{\vec{\alpha}_r}^{\mp} = g_{\vec{\alpha}_r}^2 V_C^{-1}\chi_{\vec{\alpha}_r}^{\pm}$, which, for a random dipole orientation, can be written with the help of Eq. (27) as

$$\gamma_{\vec{\alpha}_r}^{\mp} = \frac{\gamma_0}{4\pi}\lambda^2\left(\sum_i \hat{\alpha}_i L_i^{-1}\right)^{-1}\Big/ V_C. \tag{28}$$

Since $V_C/(\sum_i \hat{\alpha}_i L_i^{-1})^{-1}$ represents the transverse section seen by mode $\vec{\alpha}$, $\lambda^2(\sum_i \hat{\alpha}_i L_i^{-1})^{-1}/V_C$ is the diffraction solid angle of mode $\vec{\alpha}$. Then, relation (28) has a simple interpretation: the damping

coefficient $\gamma_{\vec{\alpha}}$ is given by the product of γ_0 with the ratio between the diffraction and the total solid angle.

For a pencil-shaped volume (length L and diameter d), $\gamma_{\vec{\alpha}_r}$ reaches its maxima for axial modes $\vec{\alpha}_0$, with $\gamma_{\max} = (\gamma_0/4\pi)\lambda^2 d^{-2}$, and its minima for perpendicular modes $\vec{\alpha}_p$, with $\gamma_{\vec{\alpha}_{pr}} = (\gamma_0/4\pi)\lambda^2(dL)^{-1}$. Approximation (15) is certainly valid if the Fresnel number is such that $\mathfrak{F} > 1$, as discussed in Appendix B. However, we have performed the calculation for any $\mathfrak{F}$, finding for the two resonant axial modes

$$\gamma_{\max} \simeq \frac{\gamma_0}{4\pi}\frac{\lambda^2}{d^2}\frac{2}{1+(1+\mathfrak{F}^{-2})^{1/2}} .$$

This expression shows that the previous result (28) is valid for $\mathfrak{F} \gtrsim 1$, whereas for $\mathfrak{F} \ll 1$ we get $\gamma_{\max} = (\gamma_0/4\pi)\lambda/L$, in agreement with the computer calculation of Ref. 2.[13] Furthermore, we have also performed the calculation, for $\mathfrak{F} \sim 1$, for the first nonaxial resonant mode $\vec{\alpha}_{1r}$, finding $\gamma_{\vec{\alpha}_{1r}} \sim \frac{1}{2}\gamma_{\max}$, in good agreement with Eq. (28) specialized for this case.

We conclude then that Eqs. (28) and (26) are substantially valid if $\mathfrak{F}$ is not too small with respect to 1. The strong dependence of $\gamma_{\vec{\alpha}_r}$ on the α_r orientation for a pencil-shaped volume is the main mechanism which determines the direction of the emission of the superfluorescence burst.

B. Interaction pseudo-Hamiltonian

It is useful to see from Eq. (21) the pseudo-Hamiltonian which gives the coupling between atoms induced by the field. If we write Eq. (21) as

$$\dot{W}_A(t) = \sum_{\vec{\alpha}} \{\gamma_{\vec{\alpha}}[R^-_{\vec{\alpha}}, W_A(t)R^+_{\vec{\alpha}}] + \text{H.c.}\} + \frac{1}{i\hbar}[H_p, W_A(t)], \tag{29a}$$

where

$$H_p = \hbar(\Omega + \Omega')R_3 - \hbar\sum_{\vec{\alpha}} \Omega_{\vec{\alpha}} S_{\vec{\alpha}} ,$$

we can identify H_p with the pseudo-Hamiltonian of the coupled spin system. H_p is a regular Hermitian operator, since the losses of the spin system induced by the field emission are contained in the first term of Eq. (29a). If we go back from the collective variables to the single-atom spin operators, we have

$$H_p = \hbar(\Omega + \Omega')\sum_i r_{3,i} + \hbar\sum_{i\neq j} \upsilon_{ij} r_i^+ r_j^- , \tag{29b}$$

where the second quantity on the right-hand side arises from the $\Omega_{\vec{\alpha}}$ terms of Eq. (21) and represents the interaction among the atoms induced by the field.

Let us now perform the computation for υ_{ij}:

$$\upsilon_{ij} = -\sum_{\vec{\alpha}} \Omega_{\vec{\alpha}} \exp i\vec{\alpha}\vec{x}_{ij}$$
$$= P\int d^3\vec{k}\, g^2_{\vec{k}} \sum_{\vec{\alpha}} \left(\frac{f^2(\vec{k}-\vec{\alpha})}{\eta^-_{\vec{k}}} - \frac{f^2(\vec{k}+\vec{\alpha})}{\eta^+_{\vec{k}}} \right) \exp i\vec{\alpha}\cdot\vec{x}_{ij} .$$

Furthermore,

$$\sum_{\vec{\alpha}} f^2(\vec{k}\pm\vec{\alpha}) \exp(i\vec{\alpha}\vec{x}_{ij}) = h(\vec{x}_{ij}) \exp(\mp i\vec{k}\vec{x}_{ij}), \tag{30}$$

where $h(\vec{x}_{ij})$ has a maximum, $h=1$, for $\vec{x}_{ij} = \vec{x}_i - \vec{x}_j = 0$, and decreases linearly to $dL_i^{-1} \simeq 0$ for $(\vec{x}_{ij})_i = L_i$, $i = x, y, z$. [$h(\vec{x}_{ij}) = \prod_i \Lambda(\vec{x}_{ij})_i / L_i$, where the triangle functions are defined in Appendix B.] Substituting Eq. (30) into the expression for υ_{ij}, and performing the relative calculations, we have

$$\upsilon_{ij} \propto h(\vec{x}_{ij}) \frac{\cos\Omega c^{-1}|\vec{x}_{ij}|}{|\vec{x}_{ij}|} . \tag{31}$$

One can see that our pseudo-Hamiltonian H_p is the same as the one derived in Refs. 14 and 15, apart from the quantity $h(\vec{x}_{ij})$, which is 1 in the references mentioned above. In our theory, the factor $h(\vec{x}_{ij})$ comes in because we have neglected the imaginary part of $\Gamma^{\pm}_{\vec{\alpha},\vec{\alpha}'} = \int d\tau A^{\pm}_{\vec{\alpha},\vec{\alpha}'}$, with $\vec{\alpha} \neq \vec{\alpha}'$, in Eq. (17). It is easy to verify that if we leave in (17) the $\Gamma_{\vec{\alpha},\vec{\alpha}'}$ coefficients, $\vec{\alpha} \neq \vec{\alpha}'$, we have $h(\vec{x}) = 1$. However, the change brought in by having $h(\vec{x}_{ij})$ instead of 1 is very small, since it gives a sensitive reduction only to the interaction coefficients between the spins which are at the opposite edges of V_C. As it is well known, a small change in the direct interaction range should not play any role in a cooperative phenomenon, since it changes very little the cooperative range. This argument should confirm the considerations made in Appendix B about neglecting the imaginary part of $A^{\pm}_{\vec{\alpha},\vec{\alpha}'}(\tau)$.

IV. EMITTED RADIATION

To link the property of the radiated field to W_A, we have to go back to the formulation of the problem in terms of projector technique. We must obtain W_F and then calculate the field quantities. In Appendix C we give all the lengthy but straightforward mathematical calculations necessary to arrive at the field quantities.

We state here the formula for the average number of photons emitted per unit time, per unit solid angle in the $\hat{k}$ direction. The average number of photons released per unit time by the atomic system to the field mode $\vec{k}$ is given by $\langle d(a^{\dagger}_{\vec{k}} a_{\vec{k}})/dt \rangle$, so that the number of photons delivered per unit

time in the solid angle $\Delta\Omega$ is given by:

$$I_{\Delta\Omega} = \sum_{\vec{k},\hat{k}\in\Delta\Omega} \left\langle \frac{d}{dt} (a_{\vec{k}}^{\dagger} a_{\vec{k}}) \right\rangle ,$$

where the sum is limited to the $\vec{k}$ vectors whose directions are inside $\Delta\Omega$. Replacing the summation with integrals, we have that the emitted photons per unit time per unit solid angle in direction $\hat{k}$, are given by:

$$I(\hat{k}) = (2\pi)^{-3} V^{-1} \int_0^{\infty} dk \left\langle \frac{d}{dt} (a_{\vec{k}}^{\dagger} a_{\vec{k}}) \right\rangle k^2 . \qquad (32)$$

As we deduce in Appendix C, $I(\hat{k})$ can be written as

$$I(\hat{k}) = \sum_{\vec{\alpha}} \gamma(\hat{k}) f^2(k_0 \hat{k} - \vec{\alpha}) \langle S_{\vec{\alpha}}^{+} \rangle + \gamma(\hat{k}) \left(\frac{N}{2} + \langle R_3 \rangle \right) , \qquad (33)$$

where $\gamma(\hat{k})$ [$\gamma(\hat{k}) = \gamma_0/4\pi$ for a random dipole orientation] is the normal radiation fluorescence rate per unit solid angle.

Equation (33) can be simply interpreted. The second term is nothing other than the normal emission of N uncorrelated atoms, and it is isotropic for a random dipole orientation. The first term gives rise to the coherent superfluorescence burst, provided $\langle S_{\vec{\alpha}}^{+} \rangle$ is different from 0 (i.e., the superfluorescence conditions are met). The coefficients $f^2(k_0 \hat{k} - \vec{\alpha})$ represent the fraction of the contribution to radiation in the $\hat{k}$ direction of the collective variable $S_{\vec{\alpha}}^{+}$. We can see that most of the $\hat{k}$ radiation comes from the $S_{\vec{\alpha}}^{+}$ which are resonant and have $\hat{\alpha}$ close to $\hat{k}$ (inside a diffraction angle).

Let us now link the emitted radiation to the dynamics of $\langle R_3 \rangle$. Applying m.e. (21) and performing the commutation according to Eq. (4), we have for $\langle \dot{R}_3 \rangle$:

$$\langle \dot{R}_3 \rangle = -\sum_{\vec{\alpha}} \gamma_{\vec{\alpha}}^{-} \langle S_{\vec{\alpha}}^{+} \rangle - \gamma_0 \left(\frac{N}{2} + \langle R_3 \rangle \right) . \qquad (34)$$

The total emitted photon rate, obtained by integrating Eq. (33) over all directions, is given by

$$I_t = \int I(\hat{k}) d\Omega_{\hat{k}} = \sum_{\vec{\alpha}} \gamma_{\vec{\alpha}}^{-} \langle S_{\vec{\alpha}}^{+} \rangle + \gamma_0 \left(\frac{N}{2} + \langle R_3 \rangle \right) ,$$

which compared with Eq. (34) gives

$$I_t = -\langle \dot{R}_3 \rangle . \qquad (35)$$

We can write Eq. (35) as a conservation equation for the total number of excitations:

$$\sum_{\vec{k}} \langle a_{\vec{k}}^{\dagger} a_{\vec{k}} \rangle + \langle R_3 \rangle = \text{constant} . \qquad (36)$$

A similar relation would have held for the operators themselves if we had not introduced the antiresonant terms $a^{\dagger} r^{+}, a r^{-}$ in Hamiltonian (1). In fact, antiresonant terms allow virtual processes of double excitation and deexcitation which make the conservation law no longer valid for $\sum_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + R_3$, i.e., $[\sum_{\vec{k}} a_{\vec{k}}^{\dagger} a_{\vec{k}} + R_3, H] \neq 0$. However, as we have proved in Appendix C, the antiresonant terms give no contribution to the average emitted intensity in the Markoffian approximation.

V. DYNAMICS OF THE ATOMS

From Eq. (33) we see that to obtain the emitted photon rate we must first calculate $\langle R_3(t) \rangle$ and $\langle S_{\vec{\alpha}}^{+}(t) \rangle$. Since we start from an uncorrelated state, we have the following initial conditions:

$$\langle R_3(0) \rangle = \bar{N}/2, \qquad \langle S_{\vec{\alpha}}^{+}(0) \rangle = 0 \quad \text{for every } \vec{\alpha} . \qquad (37)$$

The dynamics of $\langle R_3 \rangle$ is given by Eq. (34), while the evolution of $\langle S_{\vec{\alpha}}^{+} \rangle$ can be easily derived from m.e. (21) with the help of commutators (4) and definitions (20a) and (20b). We obtain

$$\langle \dot{S}_{\vec{\alpha}}^{+} \rangle = -\langle \dot{R}_3 \rangle + \sum_{\vec{\alpha}'} \gamma_{\vec{\alpha}'}^{-} (\langle R_{\vec{\alpha}}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}'}^{-} \rangle + \text{H.c.}) + i \sum_{\vec{\alpha}'} \Omega_{\vec{\alpha}'} (\langle R_{\vec{\alpha}'}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}}^{-} \rangle - \text{H.c.}) . \qquad (38a)$$

We can rewrite Eq. (38) as

$$\langle \dot{S}_{\vec{\alpha}}^{+} \rangle = -\langle \dot{R}_3 \rangle + 2\gamma_{\vec{\alpha}}^{-} \langle R_{\vec{\alpha}}^{+} R_3 R_{\vec{\alpha}}^{-} \rangle + G_{\vec{\alpha}}^{+} , \qquad (38b)$$

with

$$G_{\vec{\alpha}}^{+} = \sum_{\vec{\alpha}' \neq \vec{\alpha}} \gamma_{\vec{\alpha}'}^{-} (\langle R_{\vec{\alpha}}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}'}^{-} \rangle + \text{H.c.}) + i \sum_{\vec{\alpha}' \neq \vec{\alpha}} \Omega_{\vec{\alpha}}^{+} (\langle R_{\vec{\alpha}'}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}}^{-} \rangle - \text{H.c.}) . \qquad (38c)$$

In Eq. (38b) we have put in evidence the role of R_3, the population difference, and we have confined in $G_{\vec{\alpha}}^{+}$ the terms giving rise to the mode-mode coupling. Like all nonlinear phenomena where mode coupling is present, the full dynamics of $G_{\vec{\alpha}}^{+}$ can be expressed through the fourth-order correlation functions, and so on. To solve the problem one has to approximate the equations of motion with a suitable decoupling proceeding. To this end, let us consider the term $\langle R_{\vec{\alpha}'}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}}^{-} \rangle$ in Eq. (38c), and rewrite it as

$$\langle R_{\vec{\alpha}'}^{+} R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}}^{-} \rangle = -\tfrac{1}{2} \langle S_{\vec{\alpha}}^{+} \rangle - \tfrac{1}{2} \langle S_{\vec{\alpha}'}^{+} \rangle - \tfrac{1}{2} (\langle R_3 \rangle + N/2) + \sum_{\substack{i,j,l \\ l \neq j \\ l \neq i}} \langle r_i^{+} r_j^{-} r_{3,l} \rangle \exp[i(\vec{\alpha}' \cdot \vec{x}_i - \vec{\alpha} \cdot \vec{x}_j) + i(\vec{\alpha} - \vec{\alpha}') \cdot \vec{x}_l)] , \qquad (39)$$

where we have used the relations $r_{3j}r_j^- = -\frac{1}{2}r_j^-$ and $r_j^+ r_{3,j} = -\frac{1}{2}r_j^+$. Identity (39) can be easily verified going to single-atom operators through Eqs. (3) and (20). If we now assume that

$$\sum_{\substack{i,j \\ i,j\neq l}} \langle r_i^+ r_j^- r_{3,l}\rangle \exp[i(\vec{\alpha}'\cdot\vec{x}_i - \vec{\alpha}\cdot\vec{x}_j)] = \text{constant with respect to } l, \tag{40a}$$

the last term on the right-hand side in (39) gives no contribution if $\vec{\alpha}\neq\vec{\alpha}$, because

$$\sum_l \exp[i(\vec{\alpha}'-\vec{\alpha})\cdot\vec{x}_l] = N\delta_{\vec{\alpha},\vec{\alpha}'}.$$

Hence we have the real quantity

$$\langle R_{\vec{\alpha}}^+ R_{3,\vec{\alpha}-\vec{\alpha}'} R_{\vec{\alpha}}^-\rangle_{\vec{\alpha}\neq\vec{\alpha}'} = -\tfrac{1}{2}\langle S_{\vec{\alpha}}^-\rangle - \tfrac{1}{2}\langle S_{\vec{\alpha}'}^-\rangle - \tfrac{1}{2}(\langle R_3\rangle + N/2).$$

Substituting the above expression in (38c), with the help of Eqs. (27) and (34), we have for $G_{\vec{\alpha}}^-$

$$G_{\vec{\alpha}}^- = -(\gamma_0 - \gamma_{\vec{\alpha}}^-)\langle S_{\vec{\alpha}}^-\rangle + \langle \dot{R}_3\rangle . \tag{40b}$$

Neglecting $\gamma_{\vec{\alpha}}^-$ with respect to γ_0 Eq. (38b) becomes

$$\langle \dot{S}_{\vec{\alpha}}^-\rangle = 2\gamma_{\vec{\alpha}}^-\langle R_{\vec{\alpha}}^+ R_3 R_{\vec{\alpha}}^-\rangle - \gamma_0\langle S_{\vec{\alpha}}^-\rangle. \tag{41}$$

To summarize, we have shown that, under condition (40a), the term in Eq. (38b) containing the mode coupling gives rise to a relaxation in the equation of motion for $\langle S_{\vec{\alpha}}^-\rangle$. We note that in a semiclassical approach, i.e., replacing atomic variables (single or collective) with c numbers, we would not have obtained the relaxation term.

A. Threshold conditions

Taking $\langle S_{\vec{\alpha}}^-\rangle = 0$ at the initial time, the problem is now to see for which modes $\vec{\alpha}$ does $\langle S_{\vec{\alpha}}^-(t)\rangle$ have an exponential growth during the motion of the system, and which condition must be met for this to happen. To this end, we replace in Eq. (41) the relation (20b), and perform the decoupling

$$\langle S_{\vec{\alpha}}^- R_3\rangle \simeq \langle S_{\vec{\alpha}}^-\rangle\langle R_3\rangle ,$$

which allows us physical insight, and gives the order of magnitude of $\langle S_{\vec{\alpha}}^-\rangle$ during the evolution process. We have

$$\langle \dot{S}_{\vec{\alpha}}^-\rangle \simeq 2\gamma_{\vec{\alpha}}^-\langle S_{\vec{\alpha}}^-\rangle\langle R_3\rangle - \gamma_0\langle S_{\vec{\alpha}}^-\rangle + 2\gamma_{\vec{\alpha}}^-\langle (R_3+N/2)R_3\rangle . \tag{42}$$

The first term on the right-hand side in Eq. (42) gives rise to an exponential increase for $S_{\vec{\alpha}}^-$, with a growing time-dependent coefficient given by $2\gamma_{\vec{\alpha}}^-\langle R_3(t)\rangle$. The second term gives a damping. The last term represents the spontaneous-emission source, which is important at $t\simeq 0$, when $\langle S_{\vec{\alpha}}^-\rangle = 0$, but becomes irrelevant if $\langle S_{\vec{\alpha}}^-\rangle$ becomes much larger than N. This is the case for some dominant modes which, after $\langle S_{\vec{\alpha}}^-\rangle$ has risen to a small value, become unstable and grow exponentially.

Since $\langle R_3\rangle$ is always decreasing, we see that a necessary condition to be met for a mode to become unstable is given by

$$2\gamma_{\vec{\alpha}}^-\langle R_3(0)\rangle \gg \gamma_0. \tag{43}$$

For unstable modes we can neglect in Eq. (42) the two last terms on the right-hand side, provided we refer to times shorter than γ_0^{-1}, and long enough to assure that $\langle S_{\vec{\alpha}}^-\rangle$ has reached a value much larger than N.

For the modes which do not verify Eq. (43), the damping is stronger than the pumping, and at most they can increase linearly with time. Starting from the initial condition $\langle R_3\rangle = \bar{N}/2$, Eq. (43) can be written, for resonant modes, as

$$\frac{\bar{N}}{V_C} \gg \frac{4\pi}{\lambda^2}\sum_i \hat{\alpha}_i L_i^{-1}, \tag{44}$$

and we can define a threshold length l_T, that must necessarily be overcome by $L_{\max}$, to have a superfluorescence burst, as

$$l_T = \frac{4\pi}{\lambda^2}\frac{V_C}{N} . \tag{45}$$

Had we started from a more general model, including nonradiative atomic relaxation, we would have found that Eqs. (44) and (45) are replaced, respectively, by

$$\frac{\bar{N}}{V_C} \gg \frac{\gamma_t}{\gamma_0}\frac{4\pi}{\lambda^2}\sum_i \hat{\alpha}_i L_i^{-1} , \tag{44'}$$

$$l_T = \frac{\gamma_t}{\gamma_0}\frac{4\pi}{\lambda^2}\frac{V_C}{\bar{N}} , \tag{45'}$$

where γ_t is the total single-atom decay constant, including the radiative part γ_0.

B. Evolution of $\langle R_3\rangle$

Let us now return to Eq. (34) for $\langle R_3\rangle$ and to Eq. (41) for $\langle S_{\vec{\alpha}}^-\rangle$, and look for a procedure for solving $\langle R_3(t)\rangle$. We proceed in a self-consistent way, i.e., we assume that for $t \ll \gamma_0^{-1}$ only some relevant modes contribute to the determination of the dynamics of $\langle R_3(t)\rangle$. Once we have solved for $\langle R_3(t)\rangle$, we can evaluate the amplitude of the modes we have disregarded to check that they effectively give no contribution.

The choice of studying the evolution for times t such that $t \ll \gamma_0^{-1}$ stems from the fact that we are looking for cooperative effects which force emission to take place on a time scale much shorter than the single-atom decay time. We begin by calculating $\langle \dot{R}_3^2\rangle$ by means of the master equation (21):

$$\langle \dot{R}_3^2 \rangle = -\sum_{\vec{\alpha}} 2\gamma_{\vec{\alpha}} \langle R_{\vec{\alpha}}^{+} R_3 R_{\vec{\alpha}}^{-} \rangle - \langle \dot{R}_3 \rangle . \tag{46}$$

As we have already said, we consider that the decay of $\langle R_3^2 \rangle$ and of $\langle R_3 \rangle$ is determined essentially by the relevant modes, which have an exponential growth. Then, from Eqs. (46) and (41) we have, labeling with $\vec{\alpha}_d$ the dominant modes,

$$\left\langle \dot{R}_3^2 + \dot{R}_3 + \sum_{\vec{\alpha}_d} \dot{S}_{\vec{\alpha}} \right\rangle \simeq 0,$$

where we have retained only the exponential increasing term for $\langle \dot{S}_{\vec{\alpha}_d} \rangle$, according to what we said before. Neglecting $\langle R_3 \rangle$ because it is N times smaller with respect to $\langle R_3^2 \rangle$, we have

$$\left\langle R_3^2 + \sum_{\vec{\alpha}_d} S_{\vec{\alpha}} \right\rangle \simeq \text{const} = \frac{\bar{N}^2}{4} . \tag{47}$$

Equation (34), when we take into account only the dominant modes, can be rewritten as

$$\langle \dot{R}_3 \rangle = -\sum_{\vec{\alpha}_d} \gamma_{\vec{\alpha}} \langle S_{\vec{\alpha}} \rangle - \gamma_0 \left(\frac{N}{2} + \langle R_3 \rangle \right). \tag{34'}$$

Let us now make the following approximation:

$$\sum_{\vec{\alpha}_d} \gamma_{\vec{\alpha}} \langle S_{\vec{\alpha}} \rangle \simeq \gamma_{\max} \sum_{\vec{\alpha}_d} \langle S_{\vec{\alpha}} \rangle , \tag{48}$$

where $\gamma_{\max}$ is the maximum value of the $\gamma_{\vec{\alpha}}$ coefficients. Statement (48) amounts to assuming that the dominant modes are very few and all have about the same $\gamma_{\vec{\alpha}}$. How close their $\gamma_{\vec{\alpha}}$ must be to $\gamma_{\max}$ will be discussed in the *a posteriori* verification of the self-consistency of our scheme.

Substituting relations (48) and (47) in Eq. (34b) we have for $\langle \dot{R}_3 \rangle$

$$\langle \dot{R}_3 \rangle = -\gamma_{\max} [(\bar{N}/2)^2 - \langle R_3^2 \rangle] - \gamma_0 [(N/2) + \langle R_3 \rangle] . \tag{49}$$

We can easily solve Eq. (49) if we neglect the quantum fluctuation of the total population, i.e., if we let

$$\langle R_3^2 \rangle - \langle R_3 \rangle^2 = 0 \tag{50}$$

and if we assume in Eq. (49) that $\bar{N} \simeq N$. In this case, as is well known in the literature, the solution of Eq. (49) is given by

$$\langle R_3 \rangle \simeq -(\bar{N}/2) \tanh[(t - \tau_m)/\tau_p] , \tag{51}$$

with

$$\tau_p = 2/\bar{N}\gamma_{\max} \; ; \quad \tau_m \simeq \tfrac{1}{2} \tau_p \log(\bar{N}\gamma_{\max}/\gamma_0).$$

We then have found that the total population stays almost constant, and suddenly drops, for $t \simeq \tau_m$, in a time interval of order τ_p.

In regard to the assumption made in going from Eq. (49) to Eq. (51), we note that the replacement in Eq. (49) of N with $\bar{N}$ does not have physical implications so long as the conditions under which we look for the solution of $\langle R_3 \rangle$ are verified. Furthermore, we are rather confident in the results obtained through the semiclassical approximation (50) since a detailed analysis, contained in Ref. 3, has shown that, at least for the one-mode model, the main features of the solution (51) are not changed if we perform in Eq. (49) approximation (50). (In the correct results of Ref. 3, there is a 10% increase in τ_m and a 20% decrease in $[(\bar{N}/2)^2 - \langle R_3^2(\tau_m) \rangle]$.)

We must now verify the self consistency of our approach. First, we must ensure that the assumption of a cooperative dynamics faster than γ_0^{-1} is verified. This means that we must have $\tau_m \ll \gamma_0^{-1}$; i.e., from Eq. (51), that

$$\bar{N}\gamma_{\max}/\gamma_0 \gg \log(\bar{N}\gamma_{\max}/\gamma_0).$$

One can see that the above inequality coincides, within a logarithmic factor, with the threshold conditions (43) or (44), so that $\tau_m \ll \gamma_0^{-1}$ is verified whenever the necessary conditions for a mode to become unstable are satisfied.

Second, for the validity of the Markoffian approximation, we must require, according to Eq. (51), that $c^{-1} L_{\max} \ll \tau_p$, or, more explicitly, that $L_{\max} \ll c/\bar{N}\gamma_{\max}$.

The above condition practically coincides with the requirement (11) implied by the Born approximation. This can be easily seen by specializing Eq. (28b) to the case $\mathfrak{F} \geq 1$, to which we limit our consideration.

Finally, we must verify that only a few collective atomic modes, all having about the same $\gamma_{\vec{\alpha}}$, determine the dynamics of the superfluorescence process. To this end, let us evaluate the ratio $\langle S_{\vec{\alpha}'} \rangle / \langle S_{\vec{\alpha}_{\max}} \rangle$ during the buildup, i.e., for $t < t_m - t_p$, where $\langle S_{\vec{\alpha}_{\max}} \rangle$ refers to the modes with $\gamma_{\vec{\alpha}} = \gamma_{\max}$. Taking into account that during this time $\langle R_3 \rangle \simeq \text{const.} = \bar{N}/2$, we can derive from Eq. (42) that

$$\frac{\langle S_{\vec{\alpha}'}(t) \rangle}{\langle S_{\vec{\alpha}_{\max}}(t) \rangle} \simeq \frac{\gamma_{\vec{\alpha}'}}{\gamma_{\max}} \frac{2\gamma_{\max}\langle R_3 \rangle - \gamma_0}{2\gamma_{\vec{\alpha}'}\langle R_3 \rangle - \gamma_0} \times \frac{\exp[2(\gamma_{\vec{\alpha}'}\langle R_3 \rangle - \gamma_0)t] - 1}{\exp[2(\gamma_{\max}\langle R_3 \rangle - \gamma_0)t] - 1} . \tag{52a}$$

Specializing Eq. (52a) for $t \simeq \tau_m$ (the extension of Eq. (52a) to τ_m is sufficiently correct for our estimate), we have

$$\frac{\langle S_{\vec{\alpha}'}(\tau_m) \rangle}{\langle S_{\vec{\alpha}_{\max}}(\tau_m) \rangle} \leq \left(\bar{N} \frac{\gamma_{\max}}{\gamma_0} \right)^{(\gamma_{\vec{\alpha}'} - \gamma_{\max})/\gamma_{\max}} . \tag{52b}$$

When the threshold condition is well verified, we have $\bar{N}\gamma_{\max}/\gamma_0 \gg 1$, and we see from (52b) that only those $\vec{\alpha}'$ such that $\gamma_{\max} - \gamma_{\vec{\alpha}'} \ll \gamma_{\max} \log^{-1}(\bar{N}\gamma_{\max}/\gamma_0)$

need to be considered.

To summarize, we can say that the superfluorescence process is characterized by the instability of some collective polarization modes and by the consequent mode competition. A few strongly self-excited modes will grow much faster than the others, and will in turn determine a fast decrease of the total population, preventing the growth of other modes.[16] The conditions for this to happen are given by

$$l_T \ll L_{\max} \ll l_c , \tag{53}$$

where the left-hand side guarantees that the cooperative decay mechanism prevails on the incoherent one, while the right-hand side ensures that the decay always takes place in a vacuum of photons, so that stimulated processes do not play any role.

The results we obtained are in qualitative agreement with the general discussion of Ernst and Stehle.[17] However, we have found it useful, when dealing with a system of excited atoms, to separate clearly the problems according to the value of $l_c/L_{\max}$. Here, the condition contained in the right-hand side of Eq. (53) and the choice of collective variables for the atomic system have allowed us to arrive at more explicit results, with respect to Ref. 17, about the threshold conditions, the shape, and the time behavior of the radiated intensity for what concerns the Markoffian superfluorescence. The problem of the non-Markoffian fluorescence has been studied by Bonifacio and Lugiato.[18] Relaxing the condition $L_{\max} \ll l_c$, they obtained oscillations in the emitted intensity for $L_{\max} \sim l_c$, and the disappearance of superfluorescence for $L_{\max} \gg l_c$. The different physical behavior of non-Markoffian superfluorescence relative to the Markoffian type is due to the relevance of stimulated emission and absorption processes when $L_{\max} \gtrsim l_c$.

VI. PENCIL-SHAPED VOLUME

We now specialize to the particularly interesting case of a pencil-shaped volume (i.e., the length L and the diameter d of V_C are such that $\mathcal{F} = d^2/\lambda L \sim 1$), which will clearly show the role of the mode competition in determining the shape of the emitted radiation. Let us suppose a random dipole orientation. We label with $\vec{\alpha}_0$ and $-\vec{\alpha}_0$ the two opposite axial modes. For simplicity, we take these modes to be resonant, i.e., $\Omega = c|\vec{\alpha}_0|$, so that the damping coefficient of the other axial modes is zero, as can be seen from Appendix B. As we already said, the $\pm\vec{\alpha}_0$ modes have the maximum value of the damping coefficient, i.e., $\gamma_{\pm\vec{\alpha}_0} = (\gamma_0/4\pi)\lambda^2/d^2$, while the first off-axis modes, labeled $\vec{\alpha}_1$, if resonant,

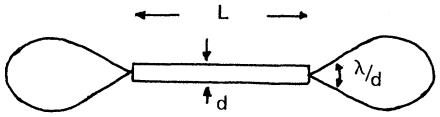

FIG. 1. Radiation pattern of the average emitted intensity from a pencil-shaped volume ($\mathcal{F} = d^2/\lambda^2 \sim 1$).

have at most a $\gamma_{\vec{\alpha}_1}$ such that $\gamma_{\vec{\alpha}_1} \leq \frac{1}{2}\gamma_{\vec{\alpha}_0}$. The γ of the second and third off-axis modes become obviously progressively smaller.

Let us make an estimate, with the help of (52b) of the ratio $\langle S_{\vec{\alpha}_1}\rangle / \langle S_{\vec{\alpha}_0}\rangle$; we have

$$\frac{\langle S_{\vec{\alpha}_1}(\tau_m)\rangle}{\langle S_{\vec{\alpha}_0}(\tau_m)\rangle} \lesssim \left(\bar{N}\frac{\gamma_{\vec{\alpha}_0}}{\gamma_0}\right)^{-1/2} . \tag{54}$$

When the threshold conditions are well verified, i.e., $\bar{N}\gamma_{\vec{\alpha}_0} \gg \gamma_0$, we notice that only the $\pm\vec{\alpha}_0$ modes practically survive. It does not matter if for increasing $\bar{N}$ the modes $\vec{\alpha}_1$ also fully satisfy Eq. (43); the mode competition will act in such a way as to progressively reduce their relative importance with respect to the axial modes. Since in the literature particular attention has been devoted to emission from a pencil-shaped active volume, let us look to the explicit formula for the radiated intensity. Taking into account that there are only the two dominant modes $\pm\vec{\alpha}_0$, we have from Eqs. (47) and (51):

$$\langle S_{\pm\vec{\alpha}_0}\rangle \simeq \frac{\bar{N}^2}{8}\,\mathrm{sech}\frac{t-\tau_m}{\tau_p} .$$

Replacing $\langle S_{\pm\vec{\alpha}_0}\rangle$ in Eq. (35), we obtain for the average number of emitted photons per unit time per unit solid angle in the $\hat{k}$ direction:

$$I(\hat{k}) \simeq \frac{\gamma_0}{4\pi}\frac{\bar{N}^2}{8} f^2\left(\frac{\Omega}{c}\hat{k} - \vec{\alpha}_0\right)\mathrm{sech}\frac{t-\tau_m}{\tau_p} + \frac{\gamma_0}{4\pi}\frac{\bar{N}^2}{8} f^2\left(\frac{\Omega}{c}\hat{k} + \vec{\alpha}_0\right)\mathrm{sech}\frac{t-\tau_m}{\tau_p}$$

+(fluorescence radiation).

As we can see in Fig. 1, the average emission of the superfluorescence burst takes place in the two "end fire modes." The radiation is all condensed around the $\pm\vec{\alpha}_0$ directions, and has a space distribution given by $f^2[(\Omega\hat{k}/c) \pm \vec{\alpha}_0]$, which implies a diffraction angle λ/d. The maximum average intensity is emitted for $t \simeq \tau_m$, when $\langle R_3\rangle = 0$. The average value of the maxima of $I(\hat{k})$ is proportional to $\bar{N}^2$, i.e., to the square of the initial population difference, while the time in which the burst occurs is given by $\tau_p \propto \bar{N}^{-1}$.

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APPENDIX A: CALCULATION FOR THE MASTER EQUATION

To obtain the evolution equation for the reduced density operator W_A, we apply the projector technique, of which we give a brief outline in the following. Let P be a projector. We can write for the total density operator W

$$W = PW + (1-P)W. \tag{A1}$$

Applying decomposition (A1) to the Liouville equation $\dot{W} = i\mathcal{L}(t)W$, where we let $\mathcal{L}$ be time dependent, we have

$$P\dot{W} = -i\mathcal{L}PW - iP\mathcal{L}[(1-P)W], \tag{A2a}$$

$$(1-P)\dot{W} = -i(1-P)\mathcal{L}[(1-P)W] - i(1-P)\mathcal{L}PW. \tag{A2b}$$

Equation (A2b) can now be integrated, treating the second term on the left-hand side as an inhomogeneity:

$$(1-P)W(t) = \exp\left[-i(1-P)\int_0^t \mathcal{L}(\tau)\,d\tau\right](1-P)W(0) - i\int_0^t d\tau \exp\left[i\int_{t-\tau}^t (1-P)\mathcal{L}(t')\,dt'\right](1-P)\mathcal{L}(t-\tau)PW(t-\tau). \tag{A3}$$

Substitution of Eq. (A3) in (A1) gives a closed non-Markoffian equation for PW:

$$P\dot{W} = -iP\mathcal{L}(t)PW(t) - i\exp\left[-i(1-P)\int_0^t \mathcal{L}(\tau)\,d\tau\right](1-P)W(0) - P\mathcal{L}(t)\int_0^t d\tau \exp\left[+i(1-P)\int_{t-\tau}^t \mathcal{L}(t')\,dt'\right](1-P)\mathcal{L}(t-\tau)PW(t-\tau). \tag{A4}$$

Define P as

$$P = |0\rangle\langle 0|\,\mathrm{tr}_F\,. \tag{A5}$$

It is easy to verify that with the initial assumptions (9) and (10) about $W(0)$, i.e., $W(0) = |0\rangle\langle 0|W_A(0)$, one has

$$(1-P)W(0) = 0, \tag{A6}$$

so that Eq. (A4) becomes homogeneous.

Starting now from Hamiltonian (5), and going to the interaction representation through the unitary operator $U = \exp[-i\hbar^{-1}(H_A + H_F)t]$, we have the following equation for $\tilde{W}$:

$$\dot{\tilde{W}} = -i\mathcal{L}(t)\tilde{W}; \quad \mathcal{L}(t) = \hbar^{-1}[\tilde{H}_{AF}, \ ] \tag{A7a}$$

with

$$\tilde{H}_{AF} = \hbar\sum_{\vec{k},\vec{\alpha}} g_{\vec{k}} V^{-1/2}[(f(\vec{k}-\vec{\alpha})\tilde{a}_{\vec{k}}\tilde{R}_{\vec{\alpha}}^{+}\exp(i\eta_{\vec{k}}^{+}t) + \mathrm{H.c.}) + (f(\vec{k}+\vec{\alpha})\tilde{a}_{\vec{k}}^{\dagger}\tilde{R}_{\vec{\alpha}}^{+}\exp(i\eta_{\vec{k}}^{+}t) + \mathrm{H.c.})], \tag{A7b}$$

where $\eta_{\vec{k}}^{\pm} = \Omega \pm \omega_{\vec{k}}$. The tilde which denotes in (A7a) and (A7b) that the operators are in the interaction representation will be dropped from now on for simplicity of notation.

Substituting Eqs. (A5) and (A7a) into Eq. (A4), we have

$$|0\rangle\langle 0|\dot{W}_A = -|0\rangle\langle 0|\int_0^t \mathrm{tr}_F\,\mathcal{L}(t)\exp\left[i(1-P)\int_{t-\tau}^t \mathcal{L}(t')\,dt'\right] \times \mathcal{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau)\,d\tau, \tag{A8}$$

where we have made use of definition (12) and of the relation $P\mathcal{L}P = 0$, which can be easily proven from Eqs. (A5) and (A7).

In the Born approximation, which is equivalent to a second-order perturbation theory, we can neglect in the integrand of (A8) the exponential factor $\exp[i(1-P)\int_{t-\tau}^t \mathcal{L}(t')\,dt']$. We must now perform the lengthy calculations to arrive to an explicit form for W_A. Substituting Eqs. (A7) in Eq. (A8) we have

$$\mathcal{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau) = \sum_{\vec{k},\vec{\alpha}} g_{\vec{k}} V^{-1/2}\{f(\vec{k}-\vec{\alpha})\{R_{\vec{\alpha}}^{+}\exp[-i\eta_{\vec{k}}^{+}(t-\tau)]|1_{\vec{k}}\rangle\langle 0|W_A(t-\tau) - \mathrm{H.c.}\} + f(\vec{k}+\vec{\alpha})\{R_{\vec{\alpha}}^{+}\exp[i\eta_{\vec{k}}^{+}(t-\tau)]|1_{\vec{k}}\rangle\langle 0|W_A(t-\tau) - \mathrm{H.c.}\}),$$

where $|1_{\vec{k}}\rangle$ is the field state characterized by a photon in the $\vec{k}$ mode. Applying $\mathrm{tr}_F\,\mathcal{L}(t)$ to the above expression, we must retain only the terms which have the diagonal projectors $|0\rangle\langle 0|$ or $|1_{\vec{k}}\rangle\langle 1_{\vec{k}}|$ on the field

space, since the trace operator over field variables wipes out all the others. We then have

$$\begin{aligned}\mathrm{tr}_F\,\mathfrak{L}(t)\mathfrak{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau) = &-\sum_{\vec{k},\vec{\alpha},\vec{\alpha}'} g_{\vec{k}}^2 V^{-1}\{f(\vec{k}-\vec{\alpha})f(\vec{k}-\vec{\alpha}')\exp(-i\eta_{\vec{k}}^{-}\tau)[R_{\vec{\alpha}'}^{-}, W_A(t-\tau)R_{\vec{\alpha}}^{+}]+\text{H.c.}\}\\ &-\sum_{\vec{k},\vec{\alpha},\vec{\alpha}'} g_{\vec{k}}^2 V^{-1}\{f(\vec{k}+\vec{\alpha})f(\vec{k}+\vec{\alpha}')\exp(i\eta_{\vec{k}}^{+}\tau)[R_{\vec{\alpha}'}^{+}, W_A(t-\tau)R_{\vec{\alpha}}^{-}]+\text{H.c.}\}\\ &+\sum_{\vec{k},\vec{\alpha},\vec{\alpha}'} g_{\vec{k}}^2 V^{-1}\{\exp i(2\Omega t-\eta_{\vec{k}}^{+}\tau)f(\vec{k}-\vec{\alpha})f(\vec{k}+\vec{\alpha}')[R_{\vec{\alpha}}^{+}, R_{\vec{\alpha}'}^{+}W_A(t-\tau)]+\text{H.c.}\}\\ &+\sum_{\vec{k},\vec{\alpha},\vec{\alpha}'} g_{\vec{k}}^2 V^{-1}\{\exp i(2\Omega t-\eta_{\vec{k}}^{-}\tau)f(\vec{k}-\vec{\alpha})f(\vec{k}+\vec{\alpha}')[R_{\vec{\alpha}'}^{+}, W_A(t-\tau)R_{\vec{\alpha}}^{+}]+\text{H.c.}\}.\end{aligned} \tag{A9}$$

Finally, replacing Eq. (A9) in Eq. (A8), one has

$$\begin{aligned}\dot{W}_A(t) = &\sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t \mathfrak{D}_{\vec{\alpha},\vec{\alpha}'}^{-}(\tau)[R_{\vec{\alpha}}^{-}, W_A(t-\tau)R_{\vec{\alpha}'}^{+}]+\text{H.c.}+\sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t \mathfrak{D}_{\vec{\alpha},\vec{\alpha}'}^{+}(\tau)[R_{\vec{\alpha}'}^{+}, W_A(t-\tau)R_{\vec{\alpha}}^{-}]+\text{H.c.}\\ &+\exp(i2\Omega t)\sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t \mathfrak{N}_{\vec{\alpha},\vec{\alpha}'}^{+}(\tau)[R_{\vec{\alpha}}^{+}, R_{\vec{\alpha}'}^{+}W_A(t-\tau)]+\text{H.c.}\\ &+\exp(i2\Omega t)\sum_{\vec{\alpha},\vec{\alpha}'}\int_0^t \mathfrak{N}_{\vec{\alpha},\vec{\alpha}'}^{-}(\tau)[R_{\vec{\alpha}'}^{+}, W_A(t-\tau)R_{\vec{\alpha}}^{-}]+\text{H.c.},\end{aligned} \tag{A10a}$$

where

$$\mathfrak{D}_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau)=\sum_{\vec{k}} g_{\vec{k}}^2 V^{-1} f(\vec{k}\pm\vec{\alpha})f(\vec{k}\pm\vec{\alpha}')\exp(\pm i\eta_{\vec{k}}^{\pm}\tau),$$

$$\mathfrak{N}_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau)=\sum_{\vec{k}} g_{\vec{k}}^2 f(\vec{k}-\vec{\alpha}')f(\vec{k}+\vec{\alpha}')\exp(-i\eta_{\vec{k}}^{\pm}\tau). \tag{A10b}$$

We see that the two last terms on the right-hand side in (A10), arising from the commutators containing R^+R^+ or R^-R^-, are affected by an intrinsic time dependence like $\exp(\pm i2\Omega t)$. Hence for times larger than Ω^{-1}, which is practically zero in our time scale, we can neglect these rapidly oscillating terms with respect to the others. Furthermore let us remark that the R^+R^+, R^-R^- terms give exactly zero contribution to the equations of motion of unphased quantities like $\langle R_3\rangle$, $\langle R^+R^-\rangle$, and so on. This approximation must not be confused with the so called rotating-wave approximation, which consists in neglecting the antiresonant terms, i.e., $a^\dagger r^+, ar^-$, in Hamiltonian (1). In fact, the antiresonant terms give rise in Eq. (A10a) not only to the disregarded rapidly varying quantities, but also to the second terms on the right-hand side with $\mathfrak{D}_{\vec{\alpha},\vec{\alpha}'}^{\pm}$, which contribution is important for the evaluation of frequency shifts.[8,19]

If we now let $V\to\infty$, the discrete field modes $\vec{k}$ go into a continuum, and we must replace $\sum_{\vec{k}}$ with $(2\pi)^{-3}V\int d^3\vec{k}_{\pm}$ obtaining from $\mathfrak{D}_{\vec{\alpha},\vec{\alpha}'}^{\pm}$ of (A10b) the coefficients $A_{\vec{\alpha},\vec{\alpha}'}^{\pm}$ of relation (13).

APPENDIX B: EVALUATION OF $A_{\vec{\alpha},\vec{\alpha}'}^{\pm}(\tau)$

We want to prove first that $A_{\vec{\alpha},\vec{\alpha}'}^{-}$, with $\vec{\alpha}=\vec{\alpha}'$, as given by expression (13), can be expressed as in Eq. (16) whenever we can perform the approximation (15). To this end, let us replace in Eq. (13) the integration variable $\vec{k}$ with $\vec{k}'=\vec{k}-\vec{\alpha}$, obtaining

$$A_{\vec{\alpha},\vec{\alpha}}^{-}(\tau)=(2\pi)^{-3}\int g_{\vec{\alpha}+\vec{k}'}^2 f^2(\vec{k}')\exp(-i\eta_{\vec{\alpha}+\vec{k}'}^{-}\tau)\,d^3\vec{k}'. \tag{B1}$$

The limits of the integration over $\vec{k}'$ are practically limited by the f function to the intervals $-2\pi L_i^{-1}<k_i'<2\pi L_i^{-1}$, as can be seen from Eq. (6d). This allows us to remove from the integral in Eq. (B1) the coefficient $g_{\vec{\alpha}+\vec{k}'}^2$, evaluated at $\vec{k}'=0$. Furthermore, we can expand the exponent $\eta_{\vec{\alpha}+\vec{k}'}^{-}\tau$ in a power series of $\vec{k}'$, i.e.,

$$\begin{aligned}\eta_{\vec{\alpha}+\vec{k}'}^{-}\tau &= [\Omega - c(\alpha^2+k'^2+2\vec{\alpha}\cdot\vec{k}')^{1/2}]\tau\\ &= (\Omega-|\alpha|)\tau - c\hat{\alpha}\cdot\vec{k}'\tau\\ &\quad -[k'^2-\tfrac{1}{4}(\hat{\alpha}\cdot\vec{k}')^2]c\tau/2|\alpha|+\cdots.\end{aligned} \tag{B2}$$

When the condition

$$c|\alpha|^{-1}k_{\max}'^2\tau\ll 1 \tag{B3}$$

is satisfied, we can retain in Eq. (B2) only the first two terms. This amounts to replacing $\vec{k}$ with $\vec{k}\cdot\hat{\alpha}$ in the exponent of (13), as can be easily verified. Whenever Eq. (B3) holds, we have from Eq. (B1) that

$$A^{\mp}_{\alpha,\pm}(\tau) = (2\pi)^{-3} g^2_{\pm} \exp(-i\eta^{\mp}_{\alpha}\tau) \times \prod_{i=1}^{3} \int \mathrm{sinc}^2(k'_i L_i/2) \exp(ic\tau k'_i \hat{\alpha}_i) dk'_i , \tag{B4}$$

where we have made use of Eq. (6d) for $f(\vec{k}')$.

Each integral in Eq. (B4) is a transform of sinc^2, and it can be easily performed with a convolution of the transforms of the sinc functions. We have:

$$(2\pi)^{-1} \int \mathrm{sinc}^2(k'_i L_i/2) \exp(ic\tau \hat{\alpha}_i k'_i) dk'_i = L_i^{-1} \Lambda(c\hat{\alpha}_i \tau/L_i), \tag{B5}$$

where the triangle function Λ is defined as:

$$\Lambda(x) = \begin{cases} 1-|x| & \text{for } |x| \leq 1 \\ 0 & \text{for } |x| > 1 . \end{cases}$$

A more compact formula for $A^{\mp}_{\alpha,\pm}$ can be obtained by replacing $\Lambda(c\hat{\alpha}_i\tau/L_i)$ with $\exp(-c\hat{\alpha}_i\tau/L_i)$, which we consider a good approximation for most cases. With this substitution, we have

$$A^{\mp}_{\alpha,\pm}(\tau) = g^2_{\pm} \exp(-i\eta^{\mp}_{\alpha}\tau) \prod_{i=1}^{3} L_i^{-1} \exp\left(-\frac{c\hat{\alpha}_i}{L_i}\tau\right), \tag{B6}$$

which is equivalent to Eqs. (16a) and (16b). The proof for A^+ is analogous. The requirements that must be met in order to perform the substitution (15) are contained in the inequality (B3). We note that the largest k' and τ to be considered in (B3), are given by $k'_{\max} \sim 2\pi L^{-1}_{\min}$ and $\tau_{\max} \sim c^{-1} L_{\max}$, where $L_{\min}$ and $L_{\max}$ are, respectively, the minimum and maximum dimensions of V_C. Furthermore, one can see from Eqs. (12) and (16) that the order of magnitude of the $|\vec{\alpha}|$ of the significant modes is given by $2\pi\lambda^{-1}$, so that we can replace Eq. (B3) with

$$\mathcal{F} = L^2_{\min}/\lambda L_{\max} \gg 1, \tag{B7}$$

where $\mathcal{F}$ is the Fresnel number. The requirement (B7) on $\mathcal{F}$ can be removed, retaining in Eq. (B2) the terms in k'^2. One obtains, in this way, integrals similar to the ones of the Fresnel diffraction that slightly modify the shape of the triangle function of Eq. (B5), as it appears from an analysis of the cumbersome problem. However, this modification does not play any role as we go toward the Markoffian limit, i.e., when one deals with $\Gamma^{\pm}_{\alpha} = \int_0^{\infty} A^{\pm}_{\alpha,\pm}\, d\tau$. In particular, we verified that, for a cylindrical geometry, $\mathrm{Re}\Gamma^{\pm}_{\alpha}$ calculated through Eq. (B6) gives a correct result down to $\mathcal{F} \sim 1$.

We shall now present some arguments to show that $A^{\pm}_{\alpha,\alpha'}$ can be disregarded when $\vec{\alpha} \neq \vec{\alpha}'$. From inspection of Eq. (13) we can immediately see that the overlap of the two functions $f(\vec{k}-\vec{\alpha})$ and $f(\vec{k}-\vec{\alpha}')$ is a condition for $A^{\mp}_{\alpha,\pm}, \neq 0$. We notice also from Eq. (6d) that the overlap practically exists only when $\vec{\alpha}$ and $\vec{\alpha}'$ are near-neighbor in the discrete $\vec{\alpha}$ space. To start, consider $\vec{\alpha}$ and $\vec{\alpha}'$ directed along $L_{\max}$ (z direction), so that for near neighbors $\alpha' = \alpha \pm \Delta$, $\Delta = 2\pi L^{-1}_{\max}$. Let us stay also around resonance, because a possible mode coupling, brought in by $A^{\mp}_{\alpha,\pm}$ ($\vec{\alpha} \neq \vec{\alpha}'$) will be more effective in this case. Since the choice of a mode being exactly resonant can always be made without loss of generality on the physical results, we take $|\alpha| = c^{-1}\Omega$. Proceeding as before for $A^{\mp}_{\alpha,\pm}$, we have

$$A^{\mp}_{\alpha,\alpha+\Delta} = \begin{cases} g^2_{\alpha} V_C^{-1} \exp(-ic\Delta\tau/2) \dfrac{\sin(L_{\max}\Delta/2)(1-c\tau/L_{\max})}{L_{\max}\Delta/2}, & 0 \leq \tau \leq c/L_{\max} \\ 0, & \tau > c/L_{\max} \end{cases} \tag{B8}$$

We plot in Fig. 2 the real and imaginary parts of $A^{\mp}_{\alpha,\alpha+\Delta}(\tau)$, and $A^{\mp}_{\alpha,\alpha}(\tau)$, which is real at resonance. We see from Fig. 2 that the real part of $A^{\mp}_{\alpha,\alpha+\Delta}$ has zero area, so when one performs the time integration in (12), its contribution is strongly reduced, becoming exactly zero in the Markoffian limit. Hence these terms can be completely disregarded in the nonunitary part (damping) of the atomic evolution. For what concerns the imaginary part, we notice that it changes sign going from Δ to $-\Delta$, giving in this way to the master equation contributions of opposite sign which cancel out.

APPENDIX C: FORMULA FOR THE EMITTED RADIATION

From Eq. (32), for the emitted number of photons per unit time per unit solid angle in the $\hat{k}$ direction, we want to prove here the explicit expression (33).

The problem we are facing is to link the expectation values of field variables to the atomic ones. To do this we have first to express W_F in terms of W_A. From definitions (8) and (A5), we can write

$$\mathrm{tr}_A(1-P)W = \mathrm{tr}_A(1-|0\rangle\langle 0|\,\mathrm{tr}_F)W = W_F - |0\rangle\langle 0|, \tag{C1}$$

where the identity $\mathrm{tr}_A \mathrm{tr}_F = 1$ has been used. Let us now replace on the left-hand side of (C1) Eq. (A3) for $(1-P)W$. Remembering Eq. (A6), we have the following expression for W_F:

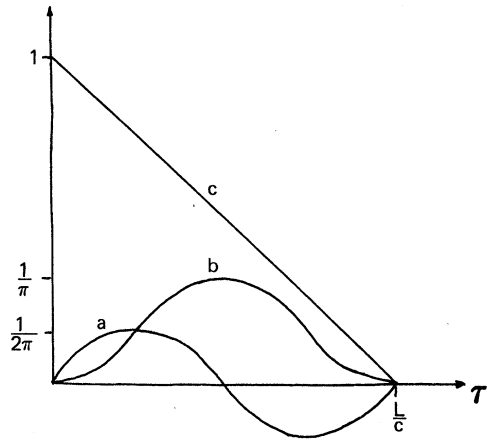

FIG. 2. Plots of (a) $\mathrm{Re}A^{-}_{\alpha,\alpha+\Delta}(\tau)$; (b) $\mathrm{Im}A^{-}_{\alpha,\alpha+\Delta}(\tau)$; (c) $A^{-}_{\alpha,\alpha}(\tau)$ (in units of $g^2_\alpha V c^{-1}$).

$$W_F = |0\rangle\langle 0| - i\,\mathrm{tr}_A \int_0^t d\tau \exp\left[i\int_{t-\tau}^t (1-P)\mathfrak{L}(t')\,dt'\right] \times (1-P)\mathfrak{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau). \tag{C2}$$

In Eq. (C2) one can now expand the exponential, retaining only the first two terms of the expansion. This is because the Born approximation we made in deriving m.e. (12) for W_A amounts to a second-order perturbation in the interaction Hamiltonian, and it would then be of no use going to a higher-order approximation in the link of W_F with W_A. Remembering also that in our case $PLP = 0$, we can rewrite Eq. (C2) as

$$W_F(t) = |0\rangle\langle 0| - i\,\mathrm{tr}_A \int_0^t \mathfrak{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau)\,d\tau + \mathrm{tr}_A \int_0^t (1-P)\int_{t-\tau}^t \mathfrak{L}(t')\,dt' \times \mathfrak{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau)\,d\tau. \tag{C3}$$

Equation (C3) links W_F to W_A in the Born approximation. Let us use it now to evaluate $\langle a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}}\rangle = \mathrm{tr}_F a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}} W_F$. Since the first and the second terms on the right-hand side of Eq. (C3) do not give any contribution, as one can easily verify, we have

$$\langle a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}}\rangle = \mathrm{tr}_A\,\mathrm{tr}_F \int_0^t a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}} \int_{t-\tau}^t \mathfrak{L}(t')\,dt' \times \mathfrak{L}(t-\tau)|0\rangle\langle 0|W_A(t-\tau)\,d\tau, \tag{C4}$$

where we have again used the fact that $\mathrm{tr}_F a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}}|0\rangle\langle 0| = 0$. With the help of Eq. (A7), we can now explicitly write Eq. (C4). The lengthy, but straightforward calculations are analogous to those performed in Appendix A. The result is:

$$\langle a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}}\rangle = 2\int_0^t d\tau \sum_{\vec{\alpha}} g^2_{\vec{k}} V^{-1}\left[f^2(\vec{k}-\vec{\alpha})\frac{\sin\eta^{-}_{\vec{k}}\tau}{\eta^{-}_{\vec{k}}}\langle R^{+}_{\vec{\alpha}} R^{-}_{\vec{\alpha}}\rangle_{t-\tau} + f^2(\vec{k}-\vec{\alpha})\frac{\sin\eta^{+}_{\vec{k}}\tau}{\eta^{+}_{\vec{k}}}\langle R^{-}_{\vec{\alpha}} R^{+}_{\vec{\alpha}}\rangle_{t-\tau}\right]. \tag{C5}$$

The time derivative of Eq. (C5), in which we are interested, takes the simpler form

$$\left\langle \frac{d}{dt}\left(a^{\dagger}_{\vec{k}} a^{-}_{\vec{k}}\right)\right\rangle = 2\int_0^t d\tau \sum_{\vec{\alpha}} g^2_{\vec{k}} V^{-1}\left[f^2(\vec{k}-\vec{\alpha})\cos\eta^{-}_{\vec{k}}\tau\langle R^{+}_{\vec{\alpha}} R^{-}_{\vec{\alpha}}\rangle_{t-\tau} + f^2(\vec{k}-\vec{\alpha})\cos\eta^{+}_{\vec{k}}\tau\langle R^{-}_{\vec{\alpha}} R^{+}_{\vec{\alpha}}\rangle_{t-\tau}\right]. \tag{C6}$$

Substituting Eq. (C6) in Eq. (32), we have for $I(\hat{k})$

$$I(\hat{k}) = \sum_{\vec{\alpha}} \int_0^t d\tau \left(\frac{2}{(2\pi)^3}\int g^2_{\vec{k}} f^2(k\hat{k}-\vec{\alpha})\cos(\eta^{-}_{\vec{k}}\tau)k^2\,dk\right)\langle R^{+}_{\vec{\alpha}} R^{-}_{\vec{\alpha}}\rangle_{t-\tau} + \sum_{\vec{\alpha}} \int_0^t d\tau \left(\frac{2}{(2\pi)^3}\int g^2_{\vec{k}} f^2(k\hat{k}-\vec{\alpha})\cos(\eta^{+}_{\vec{k}}\tau)k^2\,dk\right)\langle R^{-}_{\vec{\alpha}} R^{+}_{\vec{\alpha}}\rangle_{t-\tau}. \tag{C7}$$

Since the $\vec{k}$ integral is zero for $\tau > c^{-1}L_{\max}$, we can apply to Eq. (C7) the same considerations we did to get the Markoffian master equation, i.e., the averages $\langle R^{+}_{\vec{\alpha}} R^{-}_{\vec{\alpha}}\rangle$ and $\langle R^{-}_{\vec{\alpha}} R^{+}_{\vec{\alpha}}\rangle$, evaluated at $\tau = 0$, can be taken out of the time integrals since they have a dynamic time scale larger than $c^{-1}L_{\max}$, and the limits of the time integration can be extended from 0 to ∞. Performing the time and k integrations, we find

$$I(\hat{k}) = \sum_{\vec{\alpha}} \gamma(\hat{k}) f^2\left(\frac{\Omega}{c}\hat{k} - \vec{\alpha}\right)\langle R^{+}_{\vec{\alpha}} R^{-}_{\vec{\alpha}}\rangle, \tag{C8}$$

with $\gamma(\hat{k}) = (2\pi)^{-2}c^{-3}\Omega^2 g^2_{\Omega\hat{k}/c}$. We notice that the second term on the right-hand side of (C7) disappears, since the emission coefficient coming from $\eta^{+}_{\vec{k}}$ is exactly zero, as one can easily verify. This means that the antiresonanting terms of Hamiltonian (1), which give rise to the second term on the right-hand side of Eq. (C7), do not give any contribution to the average number of emitted photons.

From Eq. (C8) one arrives at once at Eq. (33) with the help of Eq. (20b) and the relation $\sum_{\vec{\alpha}} f^2(\Omega\hat{k}/c - \vec{\alpha}) = 1$.

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