

## Cooperative radiation processes in two-level systems: Superfluorescence\*

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We give a general nonperturbative treatment of cooperative emission in systems of  $N$  two-level atoms, starting from first principles and including inhomogeneous broadening. In particular, we study superfluorescence, which is defined as the cooperative spontaneous emission, i.e., radiation rate proportional to  $N^2$ , from an atomic system initially excited with zero macroscopic dipole moment and a uniform population difference between the excited and the fundamental states. The atomic system is described by means of collective dipole operators. A fundamental justification is given for the existence of damped "quasimodes" of the mirrorless active volume. The damping of such modes is simply due to the propagation of the Maxwell field, which escapes from the active volume. A general atom-field master equation is derived for the system atoms plus field inside the active volume, described, respectively, in terms of collective dipole operators and quasimode operators. An important feature of this equation is that inhomogeneous broadening simply appears via a time-dependent atom-field coupling constant. In this paper we give a semiclassical treatment of such a master equation. For a pencil-shaped geometry of the active volume, generalized Maxwell-Bloch equations are derived for the envelopes of the radiation inside the active volume and polarization. Such equations take into account the two directions of propagation of the radiation and the inhomogeneous broadening. Suitably phrasing our initial condition in semiclassical terms, we find that propagation effects can be neglected at all times and the generalized Maxwell-Bloch equations reduce to a simple pendulum equation. On the basis of the discussion of the pendulum equation, we conclude that superfluorescence occurs when (i) the length  $L$  of the active volume is much larger than a suitable threshold length  $L_T$  (this condition ensures that the dephasing atomic processes occur on a time scale much larger than the times characteristic of the cooperative emission); (ii) the length  $L$  is smaller or of the same order of a suitable cooperation length  $L_c$  (this condition ensures that cooperative spontaneous emission dominates stimulated processes, which give radiation proportional to  $N$ ). For  $L \ll L_c$ , one has a hyperbolic-secant superfluorescent pulse; for  $L \approx L_c$ , as one has in the recent experiments of Skribanowitz *et al.*, one finds oscillations in the cooperative decay and in the radiation emission. Such oscillations are due to the contribution of stimulated processes. For  $L \gg L_c$ , this contribution increases. As a consequence one gets more oscillations in the radiated intensity, which becomes proportional to  $N$ , so that superfluorescence effects disappear.

### I. INTRODUCTION

Cooperative emission from a system of  $N \gg 1$  atoms is a typical many-body effect, consisting in the emission of a radiation pulse with intensity proportional to  $N^2$ . A typical example is superradiance, which is a cooperative emission from a system excited by a coherent pulse into a "correlated" state which has a macroscopic electric dipole. Then the superradiant pulse is due to the emission of this dipole, and the phenomenon is describable in purely classical terms. Dicke<sup>1</sup> was the first to point out the phenomenon of superradiance, which he treated in the case of atoms confined in a volume of dimensions smaller than a wavelength. This limitation was later eliminated by Eberly and Rehler.<sup>2</sup> A basically different phenomenon is that of cooperative emission from a system of *uncorrelated* excited atoms. In this case no macroscopic dipole is initially present in the atomic system; the phenomenon is started by normal *fluorescent emission*. Later on the system spontaneously creates correlations, i.e., a macro-

scopic dipole which gives rise to a pulse whose maximum intensity is proportional to  $N^2$  and whose time duration is proportional to  $N^{-1}$ . We call this phenomenon *superfluorescence*.

In contrast to superradiance, superfluorescence is intrinsically a quantum process since, as we said, it is started by the "noise" spontaneous emission photons of ordinary fluorescence. This effect was first described in Ref. 3 in terms of a single-mode laser model.

A general quantum-mechanical approach to cooperative and noncooperative spontaneous emission has been given by Lehmberg<sup>4</sup> and Agarwal.<sup>5</sup> However, they discuss the problem only for an active volume with dimensions much smaller than a wavelength, or for very few atoms or replacing the atoms with harmonic oscillators.

In this paper we give a general nonperturbative quantum-mechanical treatment of cooperative emission in two-level systems, starting from first principles and taking inhomogeneous broadening into account. We consider an active volume with dimensions much larger than a wavelength;

in particular, we assume a pencil-shaped geometry, with maximal length  $L$ .

We distinguish between two types of superfluorescence: *pure superfluorescence*, in which the pulse has a single hyperbolic-secant shape; and *oscillatory superfluorescence*, in which the pulse exhibits oscillations. The occurrence of one or the other type of superfluorescence depends on the values of the parameters in play.

In Ref. 3 only pure superfluorescence is discussed. A treatment of pure superfluorescence from first principles is given in Ref. 6, in which a full description of the radiation pattern and of the frequency shift is obtained. In both Refs. 3 and 6 one finds that pure superfluorescence occurs when the length  $L$  of the active volume obeys the condition

$$L_T \ll L \ll L_c, \quad (1.1)$$

where  $L_c$  is the "cooperation length"<sup>3,7</sup> and, in the notation of Ref. 8,  $L_T^{-1} = \alpha$  is the "gain per unit length."

Oscillatory superfluorescence occurs when  $L_T \ll L \approx L_c$ ; these conditions are met in the recent experiments of Ref. 8. The theory we present in this paper gives a general treatment of cooperative emission, including both pure and oscillatory superfluorescence.

In Sec. II we introduce the Hamiltonian for the system atoms and field; the interaction is taken in the dipole and in the rotating-wave approximations. As in Ref. 6, we quantize the field in a volume  $V$  much larger than the active volume  $v$ . In Sec. III we define "slowly varying operators," which are the quantum analogs of the classical "envelopes."<sup>9</sup> In Sec. IV we assume a lattice structure for the active volume, and introduce collective dipole operators, whose consideration is essential for describing cooperative emission. In Sec. V we prove that inhomogeneous broadening appears in our calculation scheme as a time-dependent atom-field coupling constant.

In Sec. VI we define the quasimodes of the field *inside* the active volume (which we call internal field) and the quasimodes of the field *outside* the active volume. It turns out that the interaction Hamiltonian can be expressed completely in terms of the modes of the internal field and collective dipole operators. In Sec. VII we look at the atoms-plus-internal-field system as an open system, interacting with a bath given by the field outside the active region. We then eliminate the bath in the infinite volume limit on the quantization box  $V \rightarrow \infty$ , obtaining a damping for the modes of the internal field. In such a way we give a fundamental

meaning to the usual phenomenological introduction of a zero-temperature bath to describe the losses of the field (see, e.g., Ref. 10). In fact, the damping of the modes of the internal field in our theory is simply due to the propagation of the Maxwell field, which escapes from the active volume.

The field external to the active volume is immediately reconstructed from the internal field by the free propagation of the Maxwell field.

In Sec. VIII we give the initial conditions: no photons present and all atoms excited. This is essentially the preparation of the system performed in the experiment of Ref. 8. The analysis is specialized in Sec. IX to a pencil-shaped geometry for the active volume; it turns out that all the off-axial modes are negligible in the description of cooperative emission, when the Fresnel number is not larger than 1. In this paper we give a semiclassical treatment of the atom-field master equation for axial modes; a full quantum-mechanical treatment will be given in a separate paper.<sup>11</sup>

In Sec. X we treat the atom-field master equation in the self-consistent-field approximation. By a suitable definition of the "envelopes" of the radiation field inside the active volume and of the polarization field, we derive generalized Maxwell-Bloch equations, which take into account the two (right-left) directions of propagation for the radiation field and the inhomogeneous broadening.

When the envelopes are initially homogeneous, as is prescribed by the initial condition specified in Sec. VIII, one sees that the envelopes remain homogeneous at all times. In this situation, as is shown in Sec. XI, the Maxwell-Bloch equations reduce to equations for the two axial resonant modes.

Such equations are then proven to be equivalent to a single pendulum equation for the Bloch angle  $\varphi(t)$ . The initial condition given in Sec. VIII corresponds to the unstable equilibrium point. This drawback is due to the fact that superfluorescence is started by quantum-noise photons, which is not accounted for in our semiclassical treatment. Therefore we introduce the initial value of the Bloch angle  $\varphi(0) = (2/N)^{1/2}$  which simulates a uniform "noise" polarization. This assumption has been justified in Ref. 3(c) for the one-mode model and will be generally proven in Ref. 11.

Finally, in Secs. XII and XIII we discuss the pendulum equation with such initial condition and give a general description of superfluorescence. Our conclusion is that one has superfluorescence when the following conditions are satisfied:

$$L_T \ll L \lesssim L_c. \quad (1.2)$$

The left-hand side of the bound (1.2) has a simple

physical meaning; in fact, it is equivalent to the following condition:

$$\tau_R \ll T_2^*, \quad (1.3)$$

where  $\tau_R$  is the time duration of the pulse in the case of pure superfluorescence<sup>3</sup> and  $T_2^*$  is the reciprocal of the inhomogeneous linewidth, which is assumed to be the smallest atomic relaxation time. Then (1.3) prescribes that the time scale of cooperative emission must be much shorter than the atomic relaxation times; otherwise, the atomic decay processes would destroy cooperative emission.

The right-hand side of the bound (1.2) means that to have superfluorescence, cooperative spontaneous emission must dominate stimulated processes. In fact, a typical example of a system in which stimulated processes dominate is the usual laser, in which one has instead of (1.3),

$$L_c \ll L_T < L, \quad (1.4)$$

where  $L$  has to be considered as an effective length, if mirrors are present. The condition  $L_T < L$  prescribes that the laser is above threshold. Thus we see that the right-hand side of condition (1.2) is badly violated in the laser or in stimulated-emission amplifiers, which radiate proportionally to  $N$ . In the case  $L \ll L_c$  the pendulum becomes overdamped, stimulated processes are completely irrelevant, and we regain the results of Ref. 3. In the case  $L \approx L_c$ , which is realized in the experiment of Ref. 8, stimulated processes cause oscillations in the radiation intensity. In a separate paper<sup>12</sup> it will be shown that our treatment is in good agreement with the experimental data of Ref. 8. On the other hand, we stress that there is a big difference with the theoretical description of superfluorescence given in Ref. 8, wherein superfluorescence is treated by means of a semiclassical model of an amplifier. The conclusion of those authors is that the *only* condition for superfluorescence is  $L_T \ll L$ , i.e.,  $\alpha L \gg 1$ ; therefore any good amplifier would be superfluorescent. We claim on the contrary that the condition  $L \approx L_c$  is also necessary, because it distinguishes a superfluorescence source from a stimulated emission amplifier. We suggest two experiments to test our theory.

(i) Starting from the values of the parameters used in the experiment of Ref. 8, one could decrease the length  $L$  with the density  $\rho$  constant or decrease  $\rho$  with  $L$  constant, taking care to preserve condition  $L \gg L_T$ . In this case one should observe pure superfluorescence.

(ii) Starting from the same values of Ref. 8,

one could conversely increase the length  $L$  with fixed  $\rho$  or increase  $\rho$  with fixed  $L$ . In this case one should observe that the superfluorescent pulse becomes more and more oscillatory and broad and more and more depressed, until for  $L \gg L_c$  the superfluorescent behavior proportional to  $N^2$  completely disappears.

We mention finally that a brief account of part of the results of this paper is given in a letter<sup>6</sup> and has been presented at the 1974 Erice School of Quantum Electronics.<sup>13</sup>

## II. HAMILTONIAN

We consider  $N \gg 1$  two-level atoms, with positions  $\vec{x}_j$  and resonance frequencies  $\omega_j$  ( $j=1, \dots, N$ ), interacting through a dipole interaction with the radiation field. For the sake of simplicity we assume a random orientation of the atomic dipole moments, so that the coupling between atoms and field depends only on the modulus  $\mu$  of the atomic dipole moments, and the radiation field can be treated as a scalar field. The Hamiltonian of the system, in the rotating-wave approximation, reads

$$H = \sum_{j=1}^N \hbar \omega_j r_{3j} + \sum_{\vec{k}} \hbar c k a_{\vec{k}}^{\dagger} a_{\vec{k}} + \frac{i\hbar}{\sqrt{V}} \sum_{j=1}^N \sum_{\vec{k}} g_{\vec{k}} (a_{\vec{k}}^{\dagger} r_j^{-} e^{-i\vec{k} \cdot \vec{x}_j} - \text{H.c.}). \quad (2.1)$$

(i)  $V$  is the quantization volume of the radiation field. We take  $V$  much larger than the volume  $v$  of the region occupied by the atoms. In this way the atomic system is an open system. Irreversibility will be obtained by letting  $V \rightarrow \infty$  at the end of the calculations.

(ii)  $g_{\vec{k}}$  is the coupling constant:

$$g_{\vec{k}} = (ck\mu^2/2\hbar)^{1/2}. \quad (2.2)$$

(iii)  $r_i^{\pm}$  and  $r_{3i}$  are spin operators describing the two-level  $i$ th atom;  $a_{\vec{k}}^{\dagger}$  and  $a_{\vec{k}}$  are the field creation and annihilation operators:

$$[r_i^+, r_j^-] = 2r_{3i} \delta_{ij}, \quad [r_{3i}, r_j^{\pm}] = \pm r_i^{\pm} \delta_{ij}, \quad (2.3)$$

$$[a_{\vec{k}}, a_{\vec{k}'}^{\dagger}] = \delta_{\vec{k}\vec{k}'}. \quad (2.4)$$

$2r_{3i}$  is the population difference of the  $i$ th atom, whereas  $r_i^{\pm}$  represents the dipole of the  $i$ th atom.

The aim of the following analysis is (a) to introduce the collective dipole operators which are essential for describing coherent spontaneous emission, and (b) to eliminate the field outside the active region  $\Omega$ , obtaining then a time-evolution equation for the modes of the active region coupled to collective dipole operators. The analysis proceeds through several steps.

### III. SLOWLY-VARYING OPERATORS

In the following we shall find it convenient to describe the dynamics of the system in terms of operators which in the Schrödinger picture have an intrinsic time dependence. Specifically, the time dependence of such operators is given by

$$\begin{aligned} \tilde{B}(t) &= \exp\left(-\frac{i}{\hbar} H_0' t\right) B \exp\left(\frac{i}{\hbar} H_0' t\right), \\ H_0' &= \sum_{\vec{k}} \hbar \omega_0 a_{\vec{k}}^\dagger a_{\vec{k}} + \sum_{j=1}^N \hbar \omega_j r_{3j}, \end{aligned} \quad (3.1)$$

where  $\omega_0$  is a suitable reference frequency, which we shall specify in the following, e.g.,  $\tilde{a}_{\vec{k}}(t) = a_{\vec{k}}^\dagger \exp(i\omega_0 t)$ ,  $\tilde{r}_j^\dagger(t) = r_j^\dagger \exp(-i\omega_j t)$ ; then we consider "rotating" operators. The reason why we consider such rotating operators is that the mean value of  $\tilde{B}(t)$  varies in time much more slowly than the mean value of  $B$ , since the rotation due to the exponential factors apparent in (3.1) counteracts the fast "rotation" due to the unperturbed Hamiltonian (cf. also footnote 17).

Let  $W_S(t)$  be the density operator in the Schrödinger picture; then  $\langle \tilde{B} \rangle(t) = \text{Tr}[\tilde{B}(t)W_S(t)]$ . On the other hand, it is convenient to use a picture in which the operators  $\tilde{B}$  appear fixed in time; then we shall write

$$\langle \tilde{B} \rangle(t) = \text{Tr}[B W(t)], \quad (3.2)$$

where

$$W(t) = \exp\left(\frac{i}{\hbar} H_0' t\right) W_S(t) \exp\left(-\frac{i}{\hbar} H_0' t\right). \quad (3.3)$$

The density matrix  $W(t)$  obeys the time-evolution equation:

$$\frac{dW}{dt} = -\frac{i}{\hbar} [\tilde{H}(t), W(t)], \quad (3.4)$$

with

$$\begin{aligned} \tilde{H}(t) &= \tilde{H}_0 + \tilde{H}_I(t), \\ \tilde{H}_0 &= \sum_{\vec{k}} \hbar (ck - \omega_0) a_{\vec{k}}^\dagger a_{\vec{k}}, \\ \tilde{H}_I(t) &= \frac{i\hbar}{\sqrt{V}} \sum_{j=1}^N \sum_{\vec{k}} g_{\vec{k}}^\dagger [a_{\vec{k}}^\dagger r_j^\dagger \exp[i(\omega_0 - \omega_j)t] \\ &\quad - i\vec{k} \cdot \vec{x}_j] - \text{H.c.} \end{aligned} \quad (3.5)$$

In the following, we shall drop the tilde everywhere.

### IV. LATTICE MODEL AND COLLECTIVE DIPOLE OPERATORS

We assume that the  $N$  atoms are arranged on a rectangular regular lattice of volume  $v = L_x L_y L_z$ ; then  $N = N_x N_y N_z$  and the interatomic distances along the three axes are  $d_x = L_x/N_x$ , etc. We assume

further that  $L_x, L_y, L_z$  are much larger than the wavelengths  $\lambda_j = 2\pi c/\omega_j$ .

Let us consider the reciprocal lattice modes  $\vec{\alpha}$ :

$$\begin{aligned} \alpha_i &= (2\pi/L_i) n_i, \\ n_i &= 0, 1, \dots, N_i - 1, \quad i = x, y, z \end{aligned} \quad (4.1)$$

and the  $N$ -dimensional vectors

$$(1/\sqrt{N}) \exp(i\vec{\alpha} \cdot \vec{x}_j), \quad j = 1, 2, \dots, N,$$

which verify the following orthonormality and completeness relations:

$$\frac{1}{\sqrt{N}} \sum_j \exp[i(\vec{\alpha} - \vec{\alpha}') \cdot \vec{x}_j] = \delta_{\vec{\alpha}, \vec{\alpha}'}, \quad (4.2)$$

$$\frac{1}{N} \sum_{\vec{\alpha}} \exp[i\vec{\alpha} \cdot (\vec{x}_i - \vec{x}_j)] = \delta_{ij}. \quad (4.3)$$

We define the collective dipole operators as

$$R^\pm(\vec{\alpha}) = \sum_{j=1}^N r_j^\pm \exp(\pm i\vec{\alpha} \cdot \vec{x}_j); \quad (4.4)$$

they obey the following commutation relations:

$$[R^+(\vec{\alpha}), R^-(\vec{\alpha}')] = 2R_3(\vec{\alpha} - \vec{\alpha}'), \quad (4.5)$$

$$[R_3(\vec{\alpha}), R^\pm(\vec{\alpha}')] = \pm R^\pm(\vec{\alpha}' \pm \vec{\alpha}), \quad (4.6)$$

where

$$R_3(\vec{\alpha}) = \sum_{j=1}^N r_{3j} \exp(i\vec{\alpha} \cdot \vec{x}_j); \quad (4.7)$$

in particular, defining

$$R_3 \equiv R_3(\vec{0}) = \sum_{j=1}^N r_{3j}, \quad (4.8)$$

we have from (4.6)

$$[R_3, R^\pm(\vec{\alpha})] = \pm R^\pm(\vec{\alpha}). \quad (4.9)$$

$R_3$ , whose eigenvalues run from  $-N/2$  to  $N/2$ , is the half-population difference of the whole atomic system. We have from (4.5) and (4.6) that  $R^\pm(\vec{\alpha})$  and  $R_3$  obey angular-momentum commutation relations for each  $\vec{\alpha}$ .

Relation (4.5) can be inverted by (4.3):

$$r_j^\pm = \frac{1}{N} \sum_{\vec{\alpha}} R^\pm(\vec{\alpha}) \exp(\mp i\vec{\alpha} \cdot \vec{x}_j). \quad (4.10)$$

Then substituting (4.10) into (3.5) we get

$$H_I(t) = \frac{i\hbar}{\sqrt{V}} \sum_{\vec{\alpha}} \sum_{\vec{k}} g_{\vec{k}} [a_{\vec{k}}^\dagger R^-(\vec{\alpha}) f^*(\vec{k} - \vec{\alpha}, t) - \text{H.c.}], \quad (4.11)$$

where

$$\begin{aligned} f(\vec{\eta}, t) &= \frac{1}{N} \sum_{j=1}^N \exp(i\vec{\eta} \cdot \vec{x}_j) \exp[i(\omega_j - \omega_0)t] \\ &\equiv \langle e^{i\vec{\eta} \cdot \vec{x}} e^{i(\omega - \omega_0)t} \rangle. \end{aligned} \quad (4.12)$$

The average in (4.12) is taken over all atoms.

### V. CONTINUOUS LIMIT OVER THE FREQUENCY AND THE ATOM DISTRIBUTIONS

We assume that (i) frequencies and positions are uncorrelated, (ii) frequencies are symmetrically distributed around a central frequency. We choose  $\omega_0$  to coincide with such a central frequency. (iii) The center of the lattice lies in the origin of the reference frame.

By (i) we have

$$\begin{aligned} f(\vec{\eta}, t) &= \langle \exp(i\vec{\eta} \cdot \vec{x}) \rangle \langle \exp[i(\omega - \omega_0)t] \rangle \\ &= F(\vec{\eta}) g_1(t). \end{aligned} \quad (5.1)$$

We evaluate  $g_1(t)$  and  $F(\vec{\eta})$  in a continuous approximation. We get by (ii)

$$g_1(t) = \int_{-\infty}^{+\infty} d\xi \tilde{g}_1(\xi) e^{i\xi t}, \quad (5.2)$$

where  $\tilde{g}_1(\xi - \omega_0)$  is the normalized-to-one frequency distribution, usually a Lorentzian or a Gaussian. For the sake of simplicity we shall assume that  $g_1(t)$  is real. Further, assuming that the interatomic distance is much smaller than the wavelength  $\lambda_0 = 2\pi c/\omega_0$  of the atomic transition, we get in the continuous approximation by (iii)

$$\begin{aligned} F(\vec{\eta}) &= \frac{\int_v d^3x \rho \exp(i\vec{\eta} \cdot \vec{x})}{\int_v d^3x \rho} \\ &= \prod_{i=x,y,z} \frac{\sin(\eta_i L_i/2)}{\eta_i L_i/2}, \end{aligned} \quad (5.3)$$

where  $\rho = N/v$ . We stress that  $F(\vec{\eta})$  is a diffraction function. The exact expression for  $F(\vec{\eta})$  (i.e., without the continuous approximation over the atomic distribution) is

$$\begin{aligned} F(\vec{\eta}) &= \frac{1}{N} \sum_{j=1}^N e^{i\vec{\eta} \cdot \vec{x}_j} \\ &= \prod_{i=x,y,z} \frac{\sin(\eta_i L_i/2)}{N_i \sin(\eta_i L_i/2N_i)}. \end{aligned}$$

This is a function periodic over a Brillouin zone. Such a periodicity is relevant for x-ray transitions; however, when  $\lambda_0$  is an optical wavelength, one can take the limit  $N_i \rightarrow \infty$  (or equivalently  $d_i \rightarrow 0$ ), obtaining (5.3). Accordingly, in such limit we let the parameters  $n_i$  in Eq. (4.1) run over the integers from  $-\infty$  to  $+\infty$ . By (4.9), (4.10), (5.1), and (5.2) we have

$$H_I(t) = \frac{i\hbar}{\sqrt{V}} \sum_{\vec{\alpha}} \sum_{\vec{k}} g_{\vec{k}}(t) [a_{\vec{k}}^\dagger R^-(\vec{\alpha}) F(\vec{k} - \vec{\alpha}) - \text{H.c.}], \quad (5.4)$$

with  $g_{\vec{k}}(t) = g_k g_1(t)$ . In the limit case of homogeneous broadening  $\tilde{g}_1(\xi) = \delta(\xi)$  so that  $g_1(t) = 1$ : therefore the only complication brought by inhomogeneous broadening in our treatment is the appearance of a time-dependent coupling constant. We remark

that the coupling between the field modes  $\vec{k}$  and the atomic modes  $\vec{\alpha}$  takes place through the diffraction function  $F(\vec{k} - \vec{\alpha})$ , i.e., mode  $\vec{\alpha}$  is coupled only to modes  $\vec{k}$  lying in a diffraction angle around  $\vec{\alpha}$ . Moreover, since  $(\eta_i L_i/2)^{-1} \sin(\eta_i L_i/2)$  ( $i = x, y, z$ ) becomes  $\delta$ -like for  $L_i$  of macroscopic size and  $g_k$  varies slowly with  $k$ , we can replace  $g_{\vec{k}}(t)$  by  $g_{\vec{\alpha}}(t)$  in (5.4) obtaining

$$H_I(t) = \frac{i\hbar}{\sqrt{V}} \sum_{\vec{\alpha}} \sum_{\vec{k}} g_{\vec{\alpha}}(t) [a_{\vec{k}}^\dagger R^-(\vec{\alpha}) F(\vec{k} - \vec{\alpha}) - \text{H.c.}] \quad (5.5)$$

### VI. THE INTERNAL FIELD

Let us consider a subdivision of the quantization volume  $V$  into regions of equal volume  $v$ , one of which coincides with the active region  $\Omega$ . We indicate by  $\Omega_{\vec{m}}^\dagger$  the rectangle  $(m_x - \frac{1}{2})L_x < x < (m_x + \frac{1}{2})L_x$  etc.; then  $\Omega_0^\dagger$  coincides with the active region  $\Omega$ . Let  $\chi_{\vec{m}}(\vec{x})$  be the characteristic function of the region  $\Omega_{\vec{m}}^\dagger$ , i.e.,

$$\chi_{\vec{m}}(\vec{x}) = \begin{cases} 1, & \vec{x} \text{ inside } \Omega_{\vec{m}}^\dagger \\ 0, & \vec{x} \text{ outside } \Omega_{\vec{m}}^\dagger \end{cases}$$

The set of functions

$$\begin{aligned} u_{\vec{m}, \vec{\alpha}}^\dagger(\vec{x}) &= \chi_{\vec{m}}(\vec{x}) (1/\sqrt{v}) \exp(i\vec{\alpha} \cdot \vec{x}), \\ \alpha_i &= (2\pi/L_i)n_i, \quad n_i = 0, \pm 1, \pm 2, \dots, \quad i = x, y, z \end{aligned} \quad (6.1)$$

is a complete orthonormal set in the Hilbert space of square summable functions in the quantization volume  $V$ . Let  $E(\vec{x})$  be the positive frequency part of the field at time  $t=0$ , i.e.,

$$E(\vec{x}) = \frac{1}{\sqrt{V}} \sum_{\vec{k}} e^{i\vec{k} \cdot \vec{x}} a_{\vec{k}}^\dagger. \quad (6.2)$$

We can expand  $E(\vec{x})$  in the series of the functions  $u_{\vec{m}, \vec{\alpha}}^\dagger(\vec{x})$ :

$$E(\vec{x}) = \sum_{\vec{m}, \vec{\alpha}} \chi_{\vec{m}}(\vec{x}) \frac{1}{\sqrt{v}} e^{i\vec{\alpha} \cdot \vec{x}} A_{\vec{m}}^\dagger(\vec{\alpha}), \quad (6.3)$$

where

$$A_{\vec{m}}^\dagger(\vec{\alpha}) = \int_v d^3x \chi_{\vec{m}}(\vec{x}) \frac{1}{\sqrt{v}} e^{i\vec{\alpha} \cdot \vec{x}} E(\vec{x}). \quad (6.4)$$

From (6.4) and the orthonormality of  $u_{\vec{m}, \vec{\alpha}}^\dagger(\vec{x})$ , one obtains immediately that  $A_{\vec{m}}^\dagger(\vec{\alpha})$  obey Bose commutation relations:

$$[A_{\vec{m}}^\dagger(\vec{\alpha}), A_{\vec{m}'}^\dagger(\vec{\alpha}')] = \delta_{\vec{m}, \vec{m}'} \delta_{\vec{\alpha}, \vec{\alpha}'}. \quad (6.5)$$

Moreover, from definition (6.3) we have that the field inside the active region  $\mathcal{E}(\vec{x})$  which we shall term "internal field," is given by

$$\mathcal{E}(\vec{x}) = \frac{1}{\sqrt{v}} \sum_{\vec{\alpha}} e^{i\vec{\alpha} \cdot \vec{x}} A_0^\dagger(\vec{\alpha}). \quad (6.6)$$

Therefore  $A_0^\dagger(\vec{\alpha})$  are the modes of the internal

field, whereas  $A_{\vec{m}}^{\dagger}(\vec{\alpha})$  with  $\vec{m} \neq \vec{0}$  are the modes of the field outside  $\Omega$ . More precisely,  $A_{\vec{m}}^{\dagger}(\vec{\alpha})$  are quasimodes because they have an intrinsic line-width; nonetheless, for simplicity we shall term them "modes" in the following. Note that  $\mathcal{E}(\vec{x})$  is a periodic function with periodicity  $\vec{L}$  which coincides with  $E(\vec{x})$  only for  $\vec{x}$  inside  $\Omega_0$ . By (6.2) and (6.4) we have

$$A_{\vec{m}}^{\dagger}(\vec{\alpha}) = \left(\frac{v}{V}\right)^{1/2} \sum_{\vec{k}} a_{\vec{k}} \prod_{j=x,y,z} \exp[i(k_j - \alpha_j)m_j L_j] \times \frac{\sin[(k_j - \alpha_j)L_j/2]}{(k_j - \alpha_j)L_j/2}, \quad (6.7)$$

and conversely

$$a_{\vec{k}} = \left(\frac{v}{V}\right)^{1/2} \sum_{\vec{m}, \vec{\alpha}} A_{\vec{m}}^{\dagger}(\vec{\alpha}) \prod_{j=x,y,z} \exp[i(\alpha_j - k_j)m_j L_j] \times \frac{\sin[(k_j - \alpha_j)L_j/2]}{(k_j - \alpha_j)L_j/2}. \quad (6.8)$$

In particular, for the modes  $A_0^{\dagger}(\vec{\alpha})$  of the internal field, we obtain

$$A_0^{\dagger}(\vec{\alpha}) = \left(\frac{v}{V}\right)^{1/2} \sum_{\vec{k}} a_{\vec{k}} F(\vec{k} - \vec{\alpha}). \quad (6.9)$$

Then we get from (5.5)

$$H_I(t) = \frac{i\hbar}{\sqrt{v}} \sum_{\vec{\alpha}} g_{\alpha}(t) [A_0^{\dagger}(\vec{\alpha}) R^-(\vec{\alpha}) - \text{H.c.}]. \quad (6.10)$$

We stress that in the interaction Hamiltonian only the modes of the internal field appear. Such modes are superpositions of modes  $\vec{k}$ , such that only  $\vec{k}$  modes contained in a diffraction angle around  $\vec{\alpha}$  contribute to the mode  $\vec{\alpha}$ . By (6.10) the atomic collective mode  $R^+(\vec{\alpha})$  is coupled only with the diffraction mode  $A_0^{\dagger}(\vec{\alpha})$ . Since in the following we shall not be concerned in the modes of the field outside  $\Omega$ , we shall drop the index  $\vec{0}$  in  $A_0^{\dagger}(\vec{\alpha})$ .

## VII. ELIMINATION OF THE FIELD OUTSIDE THE ACTIVE VOLUME: THE ATOM-FIELD MASTER EQUATION

Let us (i) assume that at the initial time  $t=0$  the field outside the active region  $\Omega$  is in the vacuum state; (ii) perform the infinite volume limit  $V \rightarrow \infty$ . In such conditions one can describe in a self-contained way the dynamics of the internal field interacting with the atoms, neglecting the field outside  $\Omega$ . Such a dynamics is no longer a Hamil-

tonian one. In fact, the internal field modes are damped as a consequence of the irreversible escape (propagation) of the total Maxwell field from the active volume to the outside. Clearly this damping will be of the order of magnitude of the inverse transit times  $(L_j/c)^{-1}$ . One can picture the situation as follows: by assumption (i), the field outside  $\Omega$  can be considered as a zero-temperature bath for the system internal-field-plus-atoms; by (6.8) the free-field Hamiltonian  $\sum_{\vec{k}} \hbar(c\vec{k} - \omega_0) a_{\vec{k}}^{\dagger} a_{\vec{k}}$  introduces a linear coupling between the system and the bath; if one eliminates the bath variables in the limit  $V \rightarrow \infty$ , which makes Poincaré cycles disappear, one obtains a damped evolution for the system.

Before making these arguments quantitative, let us anticipate the result: let  $W^{(tr)}(t)$  be the density matrix for the internal-field-plus-atoms system; then  $W^{(tr)}(t)$  obeys, for  $t \geq 0$ , the self-contained evolution equation:

$$\frac{dW^{(tr)}}{dt} = -\frac{i}{\hbar} [H_F + H_I(t), W^{(tr)}(t)] + \Lambda_F W^{(tr)}(t), \quad (7.1)$$

where  $H_I(t)$  is given by Eq. (6.10) and

$$H_F = \hbar \sum_{\alpha} (c\alpha - \omega_0) A^{\dagger}(\vec{\alpha}) A(\vec{\alpha}), \quad (7.2)$$

$$\Lambda_F W = \sum_{\alpha} k(\hat{\alpha}) \{ [A(\vec{\alpha}), W A^{\dagger}(\vec{\alpha})] + \text{H.c.} \}, \quad (7.3)$$

with

$$k(\hat{\alpha}) = \frac{c}{2} \left( \frac{|\hat{\alpha}_x|}{L_x} + \frac{|\hat{\alpha}_y|}{L_y} + \frac{|\hat{\alpha}_z|}{L_z} \right), \quad \hat{\alpha} = \frac{\vec{\alpha}}{|\vec{\alpha}|}. \quad (7.4)$$

Equation (7.3) has the typical structure of the damping term of an harmonic oscillator under the influence of a zero-temperature bath,<sup>14,15</sup> and  $k^{-1}(\hat{\alpha})$  has an obvious physical meaning: It is the transit time in the active region  $\Omega$  of a photon traveling in the direction  $\hat{\alpha}$ . We shall call (7.1) the atom-field master equation (AFME). Let us now prove Eq. (7.1); the readers who are not interested in the mathematical details can skip directly to the following section.

Let us first neglect the interaction of the field with the atoms. Let  $W_F(t)$  be the density operator of the field; it obeys the equation

$$\frac{dW_F}{dt} = -\frac{i}{\hbar} \left[ \sum_{\vec{k}} (c\vec{k} - \omega_0) a_{\vec{k}}^{\dagger} a_{\vec{k}}, W_F(t) \right]. \quad (7.5)$$

By assumption (i)  $W_F(0)$  has the structure

$$W_F(0) = \sum_{m,n} \sum_{\vec{\alpha}_1, \vec{\alpha}_2, \dots, \vec{\alpha}_n} \sum_{\vec{\alpha}'_1, \vec{\alpha}'_2, \dots, \vec{\alpha}'_m} c_{mn}(\vec{\alpha}_1, \vec{\alpha}_2, \dots, \vec{\alpha}_n, \vec{\alpha}'_1, \vec{\alpha}'_2, \dots, \vec{\alpha}'_m) \times A^{\dagger}(\vec{\alpha}_1) A^{\dagger}(\vec{\alpha}_2) \cdots A^{\dagger}(\vec{\alpha}_n) |0\rangle \langle 0| A(\vec{\alpha}'_1) A(\vec{\alpha}'_2) \cdots A(\vec{\alpha}'_m).$$

In fact, this is the most general density operator which gives zero probability of finding photons outside the active region  $\Omega$ . Since we are interested only in the internal field, we consider only the expectation values of the variables

$$\mathcal{Q}_{rs} = A^\dagger(\vec{\beta}_2)A^\dagger(\vec{\beta}_2)\cdots A^\dagger(\vec{\beta}_r)A(\vec{\gamma}_1)A(\vec{\gamma}_2)\cdots A(\vec{\gamma}_s). \quad (7.6)$$

By (7.6) and (6.9) we have that such expectation values are given at time  $t$  by

$$\begin{aligned} \langle \mathcal{Q}_{rs} \rangle(t) = & \sum_{m \geq r} \sum_{n \geq s} \sum_{\vec{\alpha}_1, \vec{\alpha}_2, \dots, \vec{\alpha}_n} \sum_{\vec{\alpha}'_1, \vec{\alpha}'_2, \dots, \vec{\alpha}'_m} c_{nm}(\vec{\alpha}_1, \dots, \vec{\alpha}_n, \vec{\alpha}'_1, \dots, \vec{\alpha}'_m) \delta_{m+s-n-r,0} \\ & \times [K^*(\vec{\alpha}'_m, \vec{\beta}_1, t) K^*(\vec{\alpha}'_{m-1}, \vec{\beta}_2, t) \cdots K^*(\vec{\alpha}'_{m-r+1}, \vec{\beta}_r, t) \\ & \times K(\vec{\gamma}_s, \vec{\alpha}_1, t) K(\vec{\gamma}_{s-1}, \vec{\alpha}_2, t) \cdots K(\vec{\gamma}_1, \vec{\alpha}_s, t) \delta_{\vec{\alpha}'_{m-r}, \vec{\alpha}_{s+1}} \cdots \delta_{\vec{\alpha}'_1, \vec{\alpha}_n} \\ & + \text{other terms}], \end{aligned} \quad (7.7)$$

where

$$K(\vec{\alpha}, \vec{\alpha}', t) = \frac{v}{V} \sum_{\vec{k}} e^{-i(c\vec{k} - \omega_0)t} F(\vec{k} - \vec{\alpha}) F(\vec{k} - \vec{\alpha}') \rightarrow \frac{v}{(2\pi)^3} \int d^3k e^{-i(c\vec{k} - \omega_0)t} F(\vec{k} - \vec{\alpha}) F(\vec{k} - \vec{\alpha}'), \quad V \rightarrow \infty \quad (7.8)$$

and the "other terms" differ from the explicitly given one only by a different association of  $\vec{\alpha}'$  and  $\vec{\beta}$  as arguments of  $K^*$ ; of  $\vec{\gamma}, \vec{\alpha}$  as arguments of  $K$ ; of  $\vec{\alpha}', \vec{\alpha}$  as arguments of  $\delta$ . Let us evaluate  $K(\vec{\alpha}, \vec{\alpha}', t)$ . Since  $F(\vec{k} - \vec{\alpha})$  is sharply peaked in  $\vec{k} = \vec{\alpha}$ , we approximate  $k$  in the exponent in (7.8) by the component of  $\vec{k}$  in the  $\vec{\alpha}$  direction:

$$k - \vec{k} \cdot \hat{\alpha}, \quad \text{with } \hat{\alpha} = \vec{\alpha}/|\vec{\alpha}|.$$

Furthermore, since the contribution of terms with  $\vec{\alpha} \neq \vec{\alpha}'$  to Eqs. (7.7) and (7.18) is irrelevant, we take

$$K(\vec{\alpha}, \vec{\alpha}', t) = \delta_{\vec{\alpha}, \vec{\alpha}'} e^{-i(c\alpha - \omega_0)t} \prod_{i=x,y,z} \Gamma_i(\vec{\alpha}, t), \quad \alpha = |\vec{\alpha}| \quad (7.9)$$

where  $\Gamma_i(\vec{\alpha}, t)$  is the "triangle function." This approximation and the expression of  $\Gamma_i$  are discussed in the Appendix.

Finally, let us remark that in this way we are neglecting eventually a "geometrical" mode-mode coupling due to a partial overlapping of diffraction patterns of different modes of our finite interaction volume. On the contrary, we are not neglecting at this stage the nontrivial long-time scale mode-mode coupling induced by the nonlinear interaction with the atomic system described by Eqs. (10.2) and (10.3):

$$\Gamma_i(\vec{\alpha}, t) = \begin{cases} 0, & t \leq -L_i/c\hat{\alpha}_i, \\ (c\hat{\alpha}_i/L_i)t + 1, & -L_i/c\hat{\alpha}_i \leq t \leq 0, \\ -(c\hat{\alpha}_i/L_i)t + 1, & 0 \leq t \leq L_i/c\hat{\alpha}_i, \\ 0, & t > L_i/c\hat{\alpha}_i. \end{cases} \quad (7.9')$$

Approximating the triangle by an exponential with a decay time  $2L_i/c\hat{\alpha}_i$ ,<sup>16</sup> we get

$$K(\vec{\alpha}, \vec{\alpha}', t) = \delta_{\vec{\alpha}, \vec{\alpha}'} e^{-i(c\alpha - \omega_0)t} e^{-k(\hat{\alpha})t}, \quad (7.10)$$

where  $k(\hat{\alpha})$  is given by (7.4). Then from (7.7) we have for  $t \geq 0$

$$\begin{aligned} \langle \mathcal{Q}_{rs} \rangle(t) = & e^{i(c\beta_1 - \omega_0)t} e^{i(c\beta_2 - \omega_0)t} \cdots \\ & \times e^{i(c\beta_r - \omega_0)t} e^{-i(c\gamma_s - \omega_0)t} \cdots \\ & \times e^{-i(c\gamma_1 - \omega_0)t} e^{-k(\hat{\beta}_1)t} \cdots \\ & \times e^{-k(\hat{\beta}_r)t} e^{-k(\hat{\gamma}_s)t} \cdots e^{-k(\hat{\gamma}_1)t} \langle \mathcal{Q}_{rs} \rangle(0). \end{aligned} \quad (7.11)$$

One verifies easily that (7.11) is precisely the time evolution prescribed by the following equation for the density matrix  $W_F^{(tr)}$  of the internal field alone:

$$\frac{dW_F^{(tr)}}{dt} = -iL_F W^{(tr)}(t) + \Lambda_F W^{(tr)}(t), \quad t \geq 0, \quad (7.12)$$

where  $L_F W = \hbar^{-1}[H_F, W]$  and  $H_F$  and  $\Lambda_F$  have been defined in (7.2) and (7.3). In fact, one has

$$\begin{aligned} \langle \mathcal{Q}_{rs} \rangle(t) = & \text{Tr}\{\mathcal{Q}_{rs} W^{(tr)}(t)\} \\ = & \text{Tr}\{\mathcal{Q}_{rs} e^{-i(L_F + i\Lambda_F)t} W^{(tr)}(0)\}, \end{aligned} \quad (7.13)$$

$$W^{(tr)}(0) = W_F(0).$$

From (7.13) one easily gets (7.11) taking into account that  $L_F$  and  $\Lambda_F$  commute and using the identities

$$\begin{aligned} \text{Tr}\{A^{+m}(\vec{\alpha}) A^n(\vec{\alpha}) e^{-iL_F t} W\} = & e^{-i(n-m)(c\alpha - \omega_0)t} \\ & \times \text{Tr}\{A^{+m}(\vec{\alpha}) A^n(\vec{\alpha}) W\}, \end{aligned} \quad (7.14)$$

$$\begin{aligned} \text{Tr}\{A^{+m}(\vec{\alpha}) A^n(\vec{\alpha}) e^{\Lambda_F t} W\} = & e^{-(n+m)k(\hat{\alpha})t} \\ & \times \text{Tr}\{A^{+m}(\vec{\alpha}) A^n(\vec{\alpha}) W\}. \end{aligned} \quad (7.15)$$

Now we must prove that the "replacement" of  $[\sum_{\vec{k}} (ck - \omega_0) a_{\vec{k}}^\dagger a_{\vec{k}}, W]$  by  $L_F + i\Lambda_F$  holds also in the presence of the interaction with the atoms. This is clearly seen considering, e.g., the equation for the mean value of  $A(\vec{\alpha})$ . In fact, by (3.5) and (4.11) we have

$$\begin{aligned} \frac{d}{dt} \langle a_{\vec{k}} \rangle(t) &= -i(ck - \omega_0) \langle a_{\vec{k}} \rangle(t) \\ &+ \frac{1}{\sqrt{V}} \sum_{\vec{\alpha}'} g_{\alpha'}(t) F(\vec{k} - \vec{\alpha}') \langle R^-(\vec{\alpha}') \rangle(t). \end{aligned} \quad (7.16)$$

Integrating, one gets

$$\langle a_{\vec{k}} \rangle(t) = \exp[-i(ck - \omega_0)t] \langle a_{\vec{k}} \rangle(0) + \frac{1}{\sqrt{V}} \sum_{\vec{\alpha}'} \int_0^t dt' \exp[-i(ck - \omega_0)(t - t')] F(\vec{k} - \vec{\alpha}') \langle R^-(\vec{\alpha}') \rangle(t') g_{\alpha'}(t'), \quad (7.17)$$

and by (6.9)

$$\langle A(\vec{\alpha}) \rangle(t) = \langle A(\vec{\alpha}) \rangle^{(0)}(t) + \frac{1}{\sqrt{v}} \sum_{\vec{\alpha}'} \int_0^t K(\vec{\alpha}, \vec{\alpha}', t - t') g_{\alpha'}(t') \langle R^-(\vec{\alpha}') \rangle(t'), \quad (7.18)$$

where  $K(\vec{\alpha}, \vec{\alpha}', t)$  is given by (7.8).  $\langle A(\vec{\alpha}) \rangle^{(0)}(t)$  is the contribution of the free motion of the field, which has been analyzed previously. Then by (7.10) and (7.11) we have finally for  $t \geq 0$

$$\langle A(\vec{\alpha}) \rangle(t) = \exp[-i(c\alpha - \omega_0)t - k(\hat{\alpha})t] \langle A(\vec{\alpha}) \rangle(0) + \frac{1}{\sqrt{v}} \int_0^t \exp\{-i[c\alpha - \omega_0 - ik(\hat{\alpha})](t - t')\} g_{\alpha}(t') \langle R^-(\vec{\alpha}) \rangle(t'), \quad (7.19)$$

or equivalently

$$\frac{d}{dt} \langle A(\vec{\alpha}) \rangle(t) = -i(c\alpha - \omega_0) \langle A(\vec{\alpha}) \rangle(t) - k(\hat{\alpha}) \langle A(\vec{\alpha}) \rangle(t) + \frac{1}{\sqrt{v}} g_{\alpha}(t) \langle R^-(\vec{\alpha}) \rangle(t). \quad (7.20)$$

One verifies immediately that  $d\langle A(\vec{\alpha}) \rangle/dt$  given by Eq. (7.20) is precisely

$$\frac{d}{dt} \langle A(\vec{\alpha}) \rangle(t) = \text{Tr} \left[ A(\vec{\alpha}) \frac{d}{dt} W^{(tr)}(t) \right], \quad (7.21)$$

which concludes the proof.

Thus we have obtained irreversibility by the passage from the full description with the operators  $a_{\vec{k}}$  associated with the large ( $-\infty$ ) volume  $V$  to the contracted description with the operators  $A(\vec{\alpha})$  associated with the active volume  $v$ .

### VIII. THE INITIAL CONDITION

In the following we shall drop the label (tr) in the AFME (7.1). We consider (7.1) with the initial condition that at  $t=0$  the active region is uniformly excited with some positive population difference and no photons are present in the active region. For the sake of simplicity, we assume that all the atoms are initially excited. Typically in such initial state (i) the atoms are uncorrelated; (ii) no macroscopic polarization is present in the sample; (iii) no stimulated emission can initially occur.

This initial state, which is characterized by a negative temperature, can be obtained by incoherent pumping techniques like in a usual laser system or by a  $\pi$  pulse excitation or by the technique of Ref. 8. Because of the initial absence of a macroscopic dipole, the radiation emission can

be started only by quantum noise, which is accounted for by ordinary spontaneous emission.

### IX. PENCIL-SHAPED GEOMETRY: NEGLECT OF OFF-AXIAL MODES

The treatment of superfluorescence is simplified by assuming a pencil-shaped geometry for the active region, i.e.,

$$L_y = L_z = D, \quad L_x = L \quad \text{with } L \gg D \gg \lambda_0. \quad (9.1)$$

Then  $k(\hat{\alpha})$  in Eq. (7.4) takes the form:

$$k(\hat{\alpha}) = \frac{c}{2} \left( \frac{|\sin \vartheta| (|\cos \varphi| + |\sin \varphi|)}{D} + \frac{|\cos \vartheta|}{L} \right), \quad (9.2)$$

where  $\vartheta$  and  $\varphi$  are the polar and azimuthal angles of  $\vec{\alpha}$  with respect to the polar axes  $x$ .

Let us see how  $k(\hat{\alpha})$  changes as a function of  $\vartheta$ . For the axial modes ( $\vartheta=0, \pi$ ) we have

$$k = c/2L. \quad (9.3)$$

From the definition of  $\vec{\alpha}$  modes, we see that the first off-axial modes are tilted by an angle  $\vartheta \sim 2\pi/|\vec{\alpha}|D \sim \lambda_0/D$  from the  $x$  direction. Therefore since  $\lambda_0 \ll D$  they have a damping  $K_{\text{off}}$  such that

$$K_{\text{off}} \approx \frac{c}{2L} \left( \frac{1}{\mathfrak{X}} + 1 \right), \quad \mathfrak{X} = \frac{D^2}{L\lambda_0} \quad (9.4)$$

where  $\mathfrak{X}$  is the well-known Fresnel number, defined as the ratio between the "geometrical angle"  $D/L$  and the "diffraction angle"  $\lambda_0/D$ . We assume



that  $\mathcal{X} \sim 1$ ; then by (9.3) and (9.4) we get

$$K_{\text{off}} \sim 2k. \quad (9.5)$$

This argument quantitatively substantiates Dicke's<sup>1</sup> statement that off-axial modes are irrelevant for describing cooperative radiation processes if  $\mathcal{X} \ll 1$ . In fact, if on the one hand all modes are equally relevant to the description of normal isotropic fluorescence, and on the other hand one assumes that the cooperative and the normal processes involve two largely separated time scales  $\tau_R$  and  $\tau_0$ , i.e.,

$$\tau_R \ll \tau_0, \quad (9.6)$$

we can neglect off-axial modes in the description of superfluorescence. Then we redefine the AFME (7.1) with only the axial modes:

$$\begin{aligned} \frac{dW}{dt} &= -\frac{i}{\hbar} [H_F + H_I(t), W(t)] + \Lambda_F W(t), \\ H_F &= \sum_{\alpha} \hbar(c|\alpha| - \omega_0) A^\dagger(\alpha) A(\alpha), \\ H_I(t) &= \frac{i\hbar}{\sqrt{v}} \sum_{\alpha} g_{|\alpha|}(t) \{A^\dagger(\alpha) R^-(\alpha) - \text{H.c.}\}, \quad (9.7) \\ \Lambda_F W &= k \sum_{\alpha} \{[A(\alpha), WA^\dagger(\alpha)] + \text{H.c.}\}, \quad k = c/2L. \end{aligned}$$

The superfluorescence pulse will consist of two longitudinal diffraction modes.

#### X. SEMICLASSICAL TREATMENT: GENERALIZED MAXWELL-BLOCH EQUATIONS

In this and in the following sections we shall discuss Eqs. (9.7) in a semiclassical approximation; a fully quantum-mechanical treatment will be given in Ref. 11. In the following, for reasons of simplicity, we shall indicate the expectation value of any observable by the same symbol which denotes the quantum-mechanical observable.

From Eq. (9.7) we have in the self-consistent-field approximation:

$$\begin{aligned} \dot{A}(\alpha) &= -i(c|\alpha| - \omega_0)A(\alpha) - kA(\alpha) \\ &\quad + (g_{|\alpha|}(t)/\sqrt{v})R^-(\alpha), \quad (10.1) \end{aligned}$$

$$\dot{R}^-(\alpha) = \frac{2}{\sqrt{v}} \sum_{\alpha'} g_{|\alpha'|}(t) A(\alpha') R_3(\alpha' - \alpha), \quad (10.2)$$

$$\begin{aligned} \dot{R}_3(\alpha) &= -\frac{1}{\sqrt{v}} \sum_{\alpha'} g_{|\alpha'|}(t) [A^\dagger(\alpha') R^-(\alpha' - \alpha) \\ &\quad + A(\alpha') R^+(\alpha' + \alpha)], \quad (10.3) \end{aligned}$$

where we have taken into account Eqs. (4.5)–(4.9). Assuming that the duration of the superfluorescence pulse is much longer than the inverse of

the carrier frequency  $\omega_0$ , it is reasonable to write the internal field  $\mathcal{E}(x, t)$  given by (6.6) in the following way:

$$\mathcal{E}(x, t) = e^{ik_0 x} A_R(x, t) + e^{-ik_0 x} A_L(x, t), \quad k_0 = \omega_0/c, \quad (10.4)$$

with

$$\begin{aligned} A_R(x, t) &= \frac{1}{\sqrt{v}} \sum_{\alpha > 0} e^{i(\alpha - k_0)x} A(\alpha), \\ A_L(x, t) &= \frac{1}{\sqrt{v}} \sum_{\alpha < 0} e^{i(\alpha + k_0)x} A(\alpha), \end{aligned} \quad (10.5)$$

where the labels  $R$  and  $L$  mean "right" and "left," respectively, and  $A_{R,L}(x, t)$  are slowly varying in time and space, i.e.,<sup>17</sup>

$$\left\{ \frac{\partial}{\partial t} \right\} \{A_R, A_L\} \ll \left\{ \begin{matrix} \omega_0 \\ k_0 \end{matrix} \right\} \{A_R, A_L\}. \quad (10.6)$$

Equation (10.6) is the usual slowly-varying-envelope approximation (SVEA) (cf. Ref. 9). Similarly, defining the macroscopic polarization field

$$\mathcal{R}^-(x, t) = \frac{1}{v} \sum_{\alpha} e^{i\alpha x} R^-(\alpha), \quad (10.7)$$

we can write in analogy to (10.4)

$$\mathcal{R}^-(x, t) = e^{ik_0 x} \mathcal{R}_R^-(x, t) + e^{-ik_0 x} \mathcal{R}_L^-(x, t), \quad (10.8)$$

where the envelopes  $\mathcal{R}_{R,L}^-(x, t)$

$$\begin{aligned} \mathcal{R}_R^-(x, t) &= \frac{1}{v} \sum_{\alpha > 0} e^{i(\alpha - k_0)x} R^-(\alpha), \\ \mathcal{R}_L^-(x, t) &= \frac{1}{v} \sum_{\alpha < 0} e^{i(\alpha + k_0)x} R^-(\alpha) \end{aligned} \quad (10.9)$$

are slowly varying in space and time. Finally, let us define a field for inversion of population:

$$\mathcal{R}_3(x, t) = \frac{1}{v} \sum_{\alpha} e^{-i\alpha x} R_3(\alpha). \quad (10.10)$$

Also  $\mathcal{R}_3(x, t)$  is a real and slowly varying field and, like  $\mathcal{E}$  and  $\mathcal{R}_i$ , is a function periodic in space with periodicity  $\bar{L}$ . Let us assume that the length  $L$  is such that two  $\alpha$  modes exist satisfying the resonance condition  $|\alpha| = k_0$ . Since  $g_{|\alpha|}(t)$  varies slowly with  $|\alpha|$ , we replace it by the constant value

$$g(t) = g_{|\alpha|=k_0}(t). \quad (10.11)$$

Then from Eqs. (4.5), (4.6), (6.6), and (10.1)–(10.10), we derive by easy calculations the following equations:

$$\left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial x} + k \right) A_R(x, t) = g(t) \mathcal{R}_R^-(x, t), \quad (10.12)$$

$$\left( \frac{\partial}{\partial t} - c \frac{\partial}{\partial x} + k \right) A_L(x, t) = g(t) \mathcal{R}_L^-(x, t), \quad (10.13)$$

$$\frac{\partial \mathcal{R}^-}{\partial t}(x, t) = 2g(t) \mathcal{E}(x, t) \mathcal{R}_3(x, t), \quad (10.14)$$

$$\frac{\partial \mathcal{R}_3}{\partial t}(x, t) = -g(t) \mathcal{E}(x, t) \mathcal{R}^+(x, t) + \text{c.c.} \quad (10.15)$$

From (10.12) and (10.13) we see that  $A_R$  and  $A_L$  propagate in the right and left directions, respectively.

Using Eqs. (10.4), (10.8), (10.14), and (10.15) and neglecting, consistently with the SVEA, terms rapidly varying on a space scale  $k_0^{-1}$ , one obtains

$$\frac{\partial \mathcal{R}_R^-}{\partial t}(x, t) = 2g(t) A_R(x, t) \mathcal{R}_3(x, t), \quad (10.16)$$

$$\frac{\partial \mathcal{R}_L^-}{\partial t}(x, t) = 2g(t) A_L(x, t) \mathcal{R}_3(x, t), \quad (10.17)$$

$$\begin{aligned} \frac{\partial \mathcal{R}_3}{\partial t}(x, t) = & -g(t) [A_R(x, t) \mathcal{R}_R^+(x, t) \\ & + A_L(x, t) \mathcal{R}_L^+(x, t) + \text{c.c.}] \end{aligned} \quad (10.18)$$

We call the set of equations (10.12), (10.13), and (10.16)–(10.18) the “generalized Maxwell-Bloch equations” (GMBE). In fact, they are quite similar to the equations derived in Ref. 9. However, the derivation given here starts from first principles and furthermore the present equations give account of both directions of propagation as well as of inhomogeneous broadening.

The field outside the active volume is the solution of the Maxwell equations in the vacuum which continuously matches the internal field on the boundary of the active volume. We stress that even when  $A_R$  and  $A_L$  depend only on time, the external field will be a pulse varying in space and time. Finally, let us remark that our simple representation of inhomogeneous broadening via a time-dependent coupling constant  $g(t)$  clearly contains the assumption that all the atoms start dephasing at some “preferred” time  $t=0$ . However we do not find any inconsistency in using it with our initial situation and for times shorter or comparable to  $T_2^*$  as long as the system radiates as  $N^2$ . On the contrary, as we shall see, our results generalize to the non-Markovian case, results already obtained in the Markovian case.

#### XI. HOMOGENEOUS ENVELOPES: EQUATIONS FOR RESONANT MODES. THE PENDULUM EQUATION FOR THE BLOCH ANGLE

With the initial condition described in Sec. VIII, the field and polarization envelopes appearing in the GMBE, as well as  $\mathcal{R}_3(x)$ , are initially homogeneous. Owing to the structure of these equations, the envelopes remain homogeneous during

the time evolution. In such a situation we have from (10.5), (10.8), and (10.10)

$$\begin{aligned} A_R(x, t) &= \frac{1}{v} \int dx A_R(x, t) = \frac{1}{\sqrt{v}} A(k_0), \\ A_L(x, t) &= (1/\sqrt{v}) A(-k_0), \\ \mathcal{R}_R^-(x, t) &= (1/v) R^-(k_0), \\ \mathcal{R}_L^-(x, t) &= (1/v) R^-(k_0), \\ \mathcal{R}_3(x, t) &= (1/v) R_3. \end{aligned} \quad (11.1)$$

Then the GMBE reduce to the following equations for the resonant modes  $A(\pm k_0)$  and  $R(\pm k_0)$ :

$$\begin{aligned} \dot{A}(\pm k_0) &= -k A(\pm k_0) + [g(t)/\sqrt{v}] R^-(\pm k_0), \\ \dot{R}^-(\pm k_0) &= 2[g(t)/\sqrt{v}] A(\pm k_0) R_3, \\ \dot{R}_3 &= -[g(t)/\sqrt{v}] [A^\dagger(k_0) R^-(k_0) \\ &\quad + A^\dagger(-k_0) R^-(k_0) + \text{c.c.}]. \end{aligned} \quad (11.2)$$

Let us assume for definiteness that the frequency density  $\bar{g}_1(\xi)$  in Eq. (5.2) is Lorentzian, so that

$$g_1(t) = \exp(-|t|/2T_2^*), \quad (11.3)$$

where  $T_2^*$  is the reciprocal of the inhomogeneous linewidth. We assume that  $T_2^*$  is the smallest atomic relaxation time. We have for  $t \geq 0$

$$g(t) = g_0 \exp(-t/2T_2^*), \quad g_0 = g_{k_0}. \quad (11.4)$$

Let us define the total quantities:

$$\begin{aligned} R_T &= [R^+(k_0) R^-(k_0) + R^+(-k_0) R^-(-k_0)]^{1/2}, \\ A_T &= [A^\dagger(k_0) A(k_0) + A^\dagger(-k_0) A(-k_0)]^{1/2}. \end{aligned} \quad (11.5)$$

From Eqs. (11.2) we derive the following closed set of equations in  $R_T$ ,  $A_T$ , and  $R_3$ :

$$\frac{d}{dt} (R_T^2 + R_3^2) = 0, \quad (11.6)$$

$$\frac{d}{dt} (A_T^2 + R_3) = -2k A_T^2, \quad (11.7)$$

$$\ddot{R}_3 + (k + 1/2T_2^*) \dot{R}_3 = -(g_0^2/v) e^{-t/T_2^*} (2R_T^2 + 4A_T^2 R_3). \quad (11.8)$$

The interest in the system (11.6)–(11.8) is that all the quantities are phase independent; one can even assume the phases are random, as must be the case (see following discussions). With the initial condition described in Sec. VIII, the constant of motion (11.6) has the value

$$R_T^2 + R_3^2 = \frac{1}{4} N^2. \quad (11.9)$$

This conservation law is characteristic of cooperative radiation processes and in particular of superradiance and superfluorescence. In fact, Eq. (11.9) shows that if  $R_3$  decays from  $N/2$  to  $-N/2$ , the system develops a macroscopic polar-

ization  $R_T$  proportional to  $N$  when  $R_3 \approx 0$ , even if initially  $R_T \approx 0$ . [If not all the atoms are initially excited our initial condition and Eq. (19) show that  $N$  has to be intended everywhere as the "effective number of active atoms," i.e., the initial value of the population difference  $2\langle R_3 \rangle$ .]

By (11.9) we can introduce the Bloch angle  $\varphi(t)$  as follows:

$$\begin{aligned} R_3(t) &= (N/2) \cos \varphi(t), \\ R_T(t) &= (N/2) \sin \varphi(t). \end{aligned} \quad (11.10)$$

Substituting Eqs. (11.10) into Eqs. (11.7) and (11.8), we see that such equations are equivalent to

$$\begin{aligned} A_T(t) &= (\sqrt{v}/2g_0) \dot{\varphi}(t) \exp(t/2T_2^*), \\ \ddot{\varphi}(t) + \left(k + \frac{1}{2T_2^*}\right) \dot{\varphi}(t) - \frac{g_0^2 N}{v} e^{-t/\tau_2^*} \sin \varphi(t) &= 0. \end{aligned} \quad (11.11)$$

Equation (11.12) represents a pendulum with a friction  $k + (2T_2^*)^{-1}$  and with a frequency which decreases exponentially in time. Equation (11.11) links the radiation output to the motion of the pendulum. In fact, Eq. (11.7) is an energy balance equation, giving the following expression for the radiation intensity:

$$\begin{aligned} I(t) &= 2kA_T^2(t) \\ &= (kv/2g_0^2) \dot{\varphi}^2(t) e^{t/\tau_2^*}. \end{aligned} \quad (11.13)$$

A few comments are in order.

(i) Because of the symmetry in the exchange of  $A(k_0)$ ,  $R^-(k_0)$  with  $A(-k_0)$ ,  $R^-(-k_0)$  in Eqs. (11.2) and in the initial condition, one has

$$\begin{aligned} A^\dagger(k_0)A(k_0) &= A^\dagger(-k_0)A(-k_0) = \frac{1}{2}A_T^2, \\ R^+(k_0)R^-(k_0) &= R^+(-k_0)R^-(-k_0) = \frac{1}{2}R_T^2. \end{aligned} \quad (11.14)$$

Then the left and right diffraction lobes of the radiation output have equal intensity.

(ii) As we said, Eqs. (11.6)–(11.8) hold also when the phases of  $A(k_0)$ ,  $A(-k_0)$ ,  $R^+(k_0)$ , and  $R^-(-k_0)$  are completely random. In such a case, the radiation field has a vanishing mean value. As we shall see in the fully quantum-mechanical treatment of Ref. 11, our initial condition prescribes precisely that these phases are random, giving therefore a vanishing field amplitude at all times. In fact, the density operator for the radiation field turns out to be diagonal in the photon number representation.

(iii) As we shall see in Sec. XII, in Eq. (11.8) the term with  $R_T^2$  is responsible for cooperative spontaneous emission (hence for superfluorescence), whereas the term with  $A_T^2 R_3$  is responsible for stimulated emission and absorption.

(iv) For  $T_2^* = \infty$  the pendulum equation (11.12) coincides with that derived in Appendix B of Ref. 3(c) from the single-mode master equation, which is the starting point of the analysis of Ref. 3. We shall elaborate on this point in Ref. 11.

(v) The initial condition given in Sec. VIII corresponds to  $\varphi(0) = \dot{\varphi}(0) = 0$ ; i.e., to the unstable equilibrium point of the pendulum. This would imply that the system does not radiate. This drawback is due to the fact that our semiclassical treatment does not take into account the quantum noise which initially starts the pendulum. In Ref. 11, neglecting fluctuations, i.e., within the approximation

$$\langle R_3^2 \rangle = \langle R_3 \rangle^2, \quad (11.15)$$

we shall rederive the pendulum equation (11.12); it will turn out that, as a result of the quantum noise, the correct initial condition is<sup>18</sup>

$$\varphi(0) = (2/N)^{1/2}, \quad \frac{d\varphi}{dt}(0) = 0. \quad (11.16)$$

In other words, the quantum treatment modifies the present classical analysis associating to the initial fully excited state a quantum-noise polarization  $R_T = (N/2)^{1/2}$ , which by (11.13) leads to  $\varphi(0) = (2/N)^{1/2}$ . Therefore the Bloch vector is no longer pointing exactly to the north pole, but is very near to it; this slight displacement from the north pole makes the pendulum move in a finite time.

In conclusion the motion of the Bloch vector during superfluorescent decay is the following. It moves on a sphere of radius  $N/2$ , and its polar angle  $\varphi$  obeys a pendulum equation. On the other hand, since we do not have any information on the phase of the initial noise polarization the azimuthal angle is completely random giving  $\langle R^\pm(\pm k_0) \rangle = 0$  [as well as  $\langle A^\pm(\pm k_0) \rangle = 0$ ]. This randomness of the phases characterizes superfluorescence with respect to superradiance, in which the phases are fixed by the phase of the coherent classical field which excites the system.

## XII. DESCRIPTION OF SUPERFLUORESCENCE

In the description of superfluorescence four time scales are involved: the "dephasing" time  $T_2^*$ , the decay time  $k^{-1}$ , and the two times  $\tau_c$  and  $\tau_R$  defined as

$$\begin{aligned} \tau_c &= (g_0 \sqrt{\rho})^{-1}, \\ \tau_R &= k\tau_c^2 = k/g_0^2 \rho, \quad \rho = N/v. \end{aligned} \quad (12.1)$$

$\tau_c$  is the characteristic time by which the field

and the atomic system exchange energy: this follows from the analysis of Ref. 19, in which the case  $T_2^* = \infty$ ,  $k=0$  is considered (cf. also Sec. XII C).  $\tau_c$  coincides with the Arecchi-Courtens cooperation time. The pendulum equation (11.12) can be rewritten in terms of  $\tau_c$  as follows:

$$\frac{d^2\varphi(t)}{dt^2} + \left(k + \frac{1}{2T_2^*}\right) \frac{d\varphi(t)}{dt} - \frac{1}{\tau_c^2} e^{-t/\tau_c^*} \sin\varphi(t) = 0.$$

$\tau_R$  coincides with the duration time of the pure superfluorescence pulses described in Ref. 3 and with the "characteristic time" of the experiments of Ref. 8. In terms of the lifetime  $\tau_0$  of an isolated atom,  $\tau_R$  is expressed as

$$\tau_R = 8\pi\tau_0/\rho\lambda_0^2 L.$$

According to the relative magnitudes of  $\tau_c$  and  $k^{-1}$ , different pictures of the cooperative emission process are obtained.

#### A. Pure superfluorescence: $k\tau_c \gg 1$

In this case the photons escape so fast from the active volume that they cannot react on the atomic system; i.e., they cannot give rise to stimulated absorption and emission. In this condition, the radiation process is due completely to cooperative spontaneous emission. Mathematically, we find that the pendulum is *overdamped*; i.e., the Bloch vector swings down monotonically from the vicinity of the north pole to the south pole, where it stops at  $t = \infty$ . It does not show oscillations around the south pole, because they might originate only from a partial reabsorption of the radiation, which cannot happen because of the fast escape of the photons. The equation for the overdamped pendulum is (11.12) without the term  $d^2\varphi(t)/dt^2$ ; neglecting in Eq. (11.12)  $1/2T_2^* \ln(k + 1/2T_2^*)$  and taking (12.1) into account, we get

$$\frac{d\varphi(t)}{dt} = \frac{1}{\tau_R} e^{-t/\tau_c^*} \sin\varphi(t). \quad (12.2)$$

We stress that the neglect of the term  $\ddot{\varphi}$  in Eq. (11.12) is equivalent to the neglect of both  $\ddot{R}_3$  and  $A_T^2 R_3$  in Eq. (11.8). As we shall show in Ref. 11, the neglect of  $\ddot{R}_3$  is a Markov approximation. We see therefore that the Markov approximation and the approximation of neglecting the stimulated processes must be made together; it is meaningless to make only one of them.

Equation (12.2) can be exactly solved as follows. Define the reduced time  $\tau$  as

$$\tau = T_2^* (1 - e^{-t/\tau_c^*}); \quad (12.3)$$

$\tau$  runs from 0 to  $T_2^*$  when  $t$  goes from 0 to  $+\infty$ ; one has  $\tau \geq t$ . Equation (12.2) can be rewritten as follows:

$$\frac{d\varphi}{d\tau} = \frac{1}{\tau_R} \sin\varphi. \quad (12.2')$$

With the initial condition  $\varphi(0) = (2/N)^{+1/2}$ , (12.2') has the solution

$$\sin\varphi(\tau) = \text{sech}[(1/\tau_R)(\tau - \tau_D)], \quad \tau_D = \frac{1}{2}\tau_R \ln N. \quad (12.4)$$

The "reduced delay time"  $\tau_D$  is the reduced time at which the radiation intensity reaches its maximum. In fact, the radiation intensity is given, with (11.13), (12.1), and (12.4) by

$$I(\tau) = \frac{g_0^2 N^2}{2k\nu} \exp\left(-\frac{t}{T_2^*}\right) \text{sech}^2\left(\frac{1}{\tau_R}(\tau - \tau_D)\right). \quad (12.5)$$

We have obtained then a typical superfluorescent pulse with peak intensity proportional to  $N^2$ .

Let us now first vary  $T_2^*$ , keeping the other parameters fixed. When  $T_2^*$  is so large with respect to  $\tau_D = \tau_R \ln\sqrt{N}$  that it can be safely replaced by infinity, one has practically  $\tau \approx t$ , so that the picture in the reduced time coincides with the one in the real time. In this case one regains the results of Ref. 3;  $\tau_R$  and  $\tau_D$  are the real width and delay time of the pulse, which has a characteristic hyperbolic-secant shape. We call such situation "pure superfluorescence." Let us now decrease  $T_2^*$  to values which remain larger than  $\tau_R \ln\sqrt{N}$ . Then, as  $T_2^*$  decreases, the pulse broadens in time, and the delay time  $t_D$  at which the pulse reaches its maximum is larger than  $\tau_D$ :

$$t_D = T_2^* \ln \frac{T_2^*}{T_2^* - \tau_R \ln\sqrt{N}} > \tau_D. \quad (12.6)$$

Finally, when  $T_2^*$  becomes smaller than  $\tau_R \ln\sqrt{N}$ , the peak in the reduced time is reached at a time  $\tau > T_2^*$ ; in this case the picture in the real time is completely different from that in the reduced time and is dominated by the time-decaying exponential  $\exp(-t/T_2^*)$  apparent in Eq. (12.5).

From this discussion we can conclude that for  $k\tau_c \gg 1$  a superfluorescent pulse can be built only if the following condition is satisfied

$$\tau_R \ll T_2^*/\ln\sqrt{N}. \quad (12.7)$$

We stress that condition  $k\tau_c \gg 1$  implies  $\tau_c \ll \tau_R$ , so that from (12.7) it follows that also

$$\tau_c \ll T_2^*/\ln\sqrt{N}. \quad (12.7')$$

Equation (12.7) gives a lower bound for the length  $L$  of the active volume when the density  $\rho$  is fixed; in fact, by (12.1) we have

$$L \gg L_T = \frac{c \ln\sqrt{N}}{2g_0^2 \rho T_2^*}. \quad (12.8)$$

Let us now fix  $T_2^*$  and increase the length  $L$  from the threshold value  $L_T$ , keeping the density  $\rho$  constant, so that  $\tau_c$  remains constant. As  $L$  increases, the pulse shrinks and the delay time,  $t_D$ , decreases. Of course, approaching the critical length (cooperative length)

$$L_c = c/2g_0\sqrt{\rho}$$

condition  $k\tau_c \gg 1$  is violated and the system exhibits a new behavior which is described in Sec. XII B. The situation described by Eq. (12.2) is the one considered in Ref. 20. These authors pointed out the increase of the delay time  $t_D$  with respect to  $\tau_D$  due to the finiteness of  $T_2^*$ , i.e., inhomogeneous broadening. An increase is actually observed in the experiments of Ref. 8, but it is still larger than the one given by Eq. (12.6). Moreover, in Ref. 20 is discussed an equation equivalent to (12.2), which holds only for  $k\tau_c \gg 1$  and cannot give account of the oscillations in the radiation intensity found in the experiments,<sup>8</sup> in which one has  $k\tau_c \approx 1$ .

#### B. Oscillatory superfluorescence: $k\tau_c \approx 1$

This is the case in the experiment of Ref. 8. For such a condition, one has by (12.1) that  $\tau_R \approx \tau_c$ . Now the emitted photons can react on the atoms, so that the pulse is due not only to cooperative spontaneous emission but also to stimulated absorption and emission. The pendulum is no longer overdamped, and exhibits oscillations around the south pole. As we shall see in Ref. 11, these oscillations are a non-Markovian effect. The radiation intensity, which is proportional to  $\dot{\varphi}^2$ , also exhibits oscillations. We have to consider the full pendulum equation (11.12). The numerical solution of Eq. (11.12) with the initial condition (11.16) and its comparison with the experimental data will be given elsewhere<sup>12</sup>; we shall limit ourselves here to a qualitative illustration of the results.

Again one gets a superfluorescent pulse with peak proportional to  $N^2$ , provided

$$\tau_c \ll T_2^* \text{ or equivalently } \tau_R \ll T_2^*. \quad (12.9)$$

Apart from the factor  $\ln\sqrt{N}$ , condition (12.9) coincides with (12.7) and prescribes again that the dephasing time  $T_2^*$  must be much larger than the times which characterize the pulse. Condition (12.9) is well satisfied in the experiment.<sup>8</sup> With respect to the solution of the overdamped pendulum equation (12.2), the solution of Eq. (11.12) shows in the range  $k\tau_c \approx 1$  the following differences: (i) oscillations are present; (ii) the pulse is broader and lower; (iii) the delay time  $t_D$  is greater than (12.6). This increase adds to that due to inhomogeneous broadening.

As  $k$  is decreased (e.g., increasing the length  $L$  with  $\rho$  fixed, or enclosing the active volume between two mirrors and increasing the reflectivity from zero to one), these differences become more and more spectacular. When  $k$  becomes such that  $k\tau_c \ll 1$ , the pulse height becomes more and more depressed, until it completely loses its  $N^2$  character. This is clearly seen considering the following limit case.

#### C. No damping: homogeneous line ( $k=0, T_2^*=\infty$ )

This case, in which the pendulum has no friction, has been analyzed in Ref. 19.  $I(t)$  is a periodic elliptic function of time which for  $N \gg 1$  gives a train of pulses each with time duration  $\tau_c$ ; the time delay  $t_D$ , at which  $I(t)$  has its first maximum, is  $t_D = \frac{1}{2}\tau_c \ln N$ . Each pulse has within a good approximation a hyperbolic-secant shape; the atomic system and the field continuously and coherently exchange energy in a time  $\tau_c$ . The height of the pulse is proportional to  $N$ , so that we have *no more superfluorescence*. We stress that in this case stimulated processes and cooperative spontaneous emission are equally important. We see that for  $T_2^* = \infty$  the transition from the case presented in Sec. XII A to that of XII C entails the transition from  $\tau_R$  to  $\tau_c$  as the characteristic time of the pulse.

### XIII. CONCLUSIONS: CONDITIONS FOR SUPERFLUORESCENCE

From the analysis of Sec. XII we conclude that the conditions for superfluorescence are<sup>21</sup>

$$k^{-1} \lesssim \tau_R \ll T_2^*. \quad (13.1)$$

The right-hand side of the bound (13.1) is due to (12.7) and (12.9) and holds up to a factor  $\ln\sqrt{N}$ ; the left-hand side stems from the fact that superfluorescence disappears when  $k^{-1} \gg \tau_R$ . We remark that bounds  $k^{-1} \lesssim \tau_c$  and  $k^{-1} \lesssim \tau_R$  coincide.

Limitations (13.1) imply the expressive bound  $k^{-1} \ll T_2^*$ , or equivalently

$$L/c \ll T_2^*/2; \quad (13.2)$$

i.e., the time the emitted photons take to get from one point of the active volume to another must be much shorter than the dephasing time  $T_2^*$ . This is clearly a necessary condition for the rise of cooperation.

Condition  $\tau_R \ll T_2^*$  means that the superfluorescent system is a laser well above threshold. It corresponds to the condition of high gain  $\alpha L \gg 1$  of Ref. 8. On the other hand, condition  $k^{-1} \lesssim \tau_R$  is never satisfied in usual lasers, because the presence of mirrors has the effect of increasing the effective length of the sample. In fact, one has

$$k^{-1} \approx 2L/c(1-R), \quad 0 \leq R \leq 1, \quad (13.3)$$

where  $R$  is the reflectivity coefficient of the mirrors. Fixing the density  $\rho$ , the bounds (13.1) can be expressed as bounds on the length  $L$  of the sample

$$\frac{c}{2g_0^2 \rho T_2^*} = L_T \ll L \leq L_c = \frac{c}{2g\sqrt{\rho}}. \quad (13.4)$$

The peculiar features of the experiments<sup>8</sup> are (i) the presence of oscillations in the radiated intensity, and (ii) the increase of the delay time  $t_D$  with respect to the value  $\tau_R \ln \sqrt{N}$ . We see that the present theory is able to explain qualitatively both these features; the quantitative agreement with the experimental data will be shown in a separate paper.<sup>12</sup>

In our theory no role is played by "propagation effects," which in Ref. 8 are indicated as essential to give an account of the findings of the experiment. In fact, propagation effects are eliminated by the homogeneity of the initial condition. We stress that in Ref. 8 the authors claim that the only condition for superfluorescence to occur is  $\tau_R \ll T_2^*$ . On the contrary, we claim that this condition is not sufficient, because one has to satisfy also the left-hand side of inequality (13.1), which in turn implies the necessary condition (13.2). These further conditions are necessary to distinguish two different phenomena: strong amplification with stimulated emission and absorption ( $\tau_R \ll T_2^*, k^{-1}$ ), in which the system radiates proportionally to  $N$ , and cooperative superfluorescence, in which stimulated processes are never as effective as spontaneous emission, and in which the atoms radiate proportionally to  $N^2$ .

*Note added in manuscript.* It is very easy to include in our treatment the effect of black-body thermal radiation by just changing in a very standard way<sup>10</sup> the form of the  $\Lambda_F$  term in master equation (7.1). This modification, however, does not change substantially our picture, since we are describing a system which is going to radiate proportionally to  $N^2$ . Practically, it is enough to take into account thermal radiation at  $t=0$  by changing the initial value of  $\dot{\varphi}$  from zero (vacuum state) to a nonzero value which can be easily determined using Eq. (11.13) and obtaining  $\dot{\varphi}^2(0) \sim \bar{n}_{\text{therm}}(\omega_0)$ .

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#### APPENDIX

Let us study  $K(\vec{\alpha}, \vec{\alpha}', t)$  defined by (7.8) in the approximation  $k \rightarrow \vec{k} \cdot \vec{\alpha}$ . Changing the integration variable from  $\vec{k}$  to  $(\vec{k} - \vec{\alpha})$ , we find that  $K(\vec{\alpha}, \vec{\alpha}', t)$  becomes the product of three one-dimensional Fourier transforms:

$$K(\vec{\alpha}, \vec{\alpha}', t) = e^{-i(c\alpha - \omega_0)t} \prod_{i=x,y,z} \Gamma_i(\vec{\alpha}, \vec{\alpha}', t),$$

$$\Gamma_i(\vec{\alpha}, \vec{\alpha}', t) = \frac{L_i}{2\pi} \int dk_i e^{-i c \hat{\alpha}_i t} F(k_i) F(k_i - \Delta_i),$$

$$F(x) = \frac{\sin x}{x}; \quad \Delta_i = \alpha'_i - \alpha_i \quad (A1)$$

These integrals can be very easy to calculate using the convolution theorem and observing that the Fourier transform of  $F$  is the characteristic function  $\chi_0$ . The final result for each component  $j$  is

$$\Gamma_i(\vec{\alpha}, \vec{\alpha}', t) = e^{-i \Delta_i c \hat{\alpha}_i t/2} \frac{\sin \frac{1}{2} \Delta_i (L_i - c \hat{\alpha}_i t)}{L_i \Delta_i / 2}$$

for  $0 \leq t \leq L_i / c \hat{\alpha}_i$  (A2)

and

$$\Gamma_i = 0 \quad \text{for } t \geq L_i / c \hat{\alpha}_i.$$

This expression for  $\Delta_i \rightarrow 0$  leads to the real "triangle" function (7.9'). For  $\Delta_i \neq 0$ ,  $\Gamma_i$  is a complex function given by

$$\Gamma_i(\vec{\alpha}, \vec{\alpha}', t) = (-1)^{n_j} \left( \frac{\sin \Delta_i c \hat{\alpha}_i t}{2n_j \pi} - i \frac{\sin^2(\Delta_i c \hat{\alpha}_i t/2)}{n_j \pi} \right), \quad (A3)$$

where we have taken into account the fact that  $\Delta_i = 2n_j \pi / L$ . Let us comment on Eq. (A3). We observe the following.

(i) Only the  $\vec{\alpha}''$ 's which are nearest neighbors to  $\alpha(n_j = \pm 1)$  can be important with respect to the triangle function  $\Gamma(\vec{\alpha}, \vec{\alpha}', t)$ . Furthermore, the contributions for  $n_j$  even or odd have opposite signs.

(ii)  $\Gamma_i$ 's are zero for  $t=0$ , whereas the triangle function is 1.

(iii) The real part of (A3) has a maximum value  $1/2\pi$  and has zero area, so that its contribution averages to zero after a transit time. This is particularly evident in Eq. (7.18) in which  $K$  appears under a time integral. Hence  $\Gamma$ 's will eventually contribute via their imaginary part, which has a maximum value  $1/\pi$ . However, from Eq. (A3) we see that the imaginary part is an odd function of  $\Delta_j$ , i.e.,  $n_j$ , so that contributions for  $n_j = \pm 1$  cancel out in Eqs. (7.7) and (7.18) provided the preparation

of the system does not introduce strong asymmetries. (See the initial condition in Sec. VIII.) (Note that the real part also changes sign:  $\hat{\alpha}_i \rightarrow -\hat{\alpha}_i$ .) On the basis of these considerations we

conclude that terms with  $\vec{\alpha} \neq \vec{\alpha}'$  play an absolutely irrelevant role, if they play any, both in Eq. (7.7) and in Eq. (7.18), and in this sense we use from now on the approximation (7.9).

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<sup>10</sup>H. Haken, in *Handbuch der Physik* (Springer-Verlag,

Berlin, 1970), Vol. XXV/2c.

<sup>11</sup>R. Bonifacio and L. A. Lugiato (unpublished).

<sup>12</sup>A. Airoidi Crescentini, R. Bonifacio, and L. A. Lugiato (unpublished).

<sup>13</sup>R. Bonifacio, in *Cooperative effects*, edited by H. Haken (North-Holland, Amsterdam, 1974), p. 97.

<sup>14</sup>W. Weidlich and F. Haake, Z. Phys. 185, 30 (1965).

<sup>15</sup>R. Bonifacio and F. Haake, Z. Phys. 200, 526 (1967).

<sup>16</sup>The factor 2 arises by imposing the condition that after a time  $L_{\max}/c$  the state of the field is the vacuum state, i.e.,  $\langle A^\dagger A \rangle = 0$ . Hence we have to require  $2k = c/L$  [see Eq. (7.15)]. We can give a general definition of  $k(\hat{\alpha})$ :

$$k^{-1}(\hat{\alpha}) \equiv \int_0^t d\tau [K(\vec{\alpha}, \vec{\alpha}, \tau)]_{\alpha = \omega_0/c},$$

which in the approximation  $\vec{k} \approx \vec{k} \cdot \hat{\alpha}$  reduces to (7.4).

However, this approximation fails if the Fresnel number is appreciably smaller than 1. We are mainly interested in the case when the Fresnel number is near 1.

<sup>17</sup>The slow variation in time is ensured by the fact that we have considered *ab initio* rotating operators.

<sup>18</sup>Cf. also Appendix B of Ref. 3(c).

<sup>19</sup>(a) R. Bonifacio and G. Preparata, Phys. Rev. A 2, 336 (1970). The short-time solution ( $t \ll \tau_c$ ) of this paper has been recently extended to a wider class of initial conditions by (b) M. E. Smithers and E. Y. C. Lu, Phys. Rev. A 9, 790 (1974).

<sup>20</sup>E. Ressayre and A. Tallet, Phys. Rev. Lett. 30, 1239 (1973).

<sup>21</sup>If  $T_2^*$  does not coincide with the smallest atomic relaxation time  $\tau_{\min}$ , then one must replace  $T_2^*$  by  $\tau_{\min}$  in all the inequalities (13.1), (13.2), etc.