

## SUPERRADIANCE AND SUBRADIANCE IN THREE-LEVEL SYSTEMS

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Cooperative phenomena in spontaneous emission may exhibit two opposite features: either an increased emission rate, usually called superradiance, or a decreased emission rate, which we call subradiance. It is shown that, in a specific case of three-level atoms, symmetry considerations permit one to find realistic initial conditions such that the system spontaneously evolves toward a subradiant system.

Interatomic interference is well-known to be responsible for the appearance of superradiant emission [1]. In the case of  $N$  initially excited atoms, the interference is essentially constructive, leading to the  $N^2$  dependence of the maximum intensity of the emitted light pulse. Interatomic interference can also be destructive. When predicting, in 1954, the existence of superradiant emission, Dicke [1] already mentioned this possibility, in the example of two neutrons in a uniform magnetic field. Since that time, the spontaneous emission of a pair of two-level systems has been studied in detail and the radiative trapping which can occur when the distance between the two particles becomes smaller than the wavelength of the transition is now well understood [2]. This radiative trapping can, in particular, manifest itself in the so-called multiple scattering phenomenon. The specific role of interatomic interference in this phenomenon has been first pointed out by Barrat [3] in his study of "coherent multiple scattering"; subsequent predictions have been verified experimentally [3]. The connection between coherent multiple scattering and superradiance has been later shown by Ernst [4]. In both studies, the possibility for an assembly of  $N$  atoms, containing one excited atom, to trap one photon is clearly demonstrated. However, it should be a priori possible that as much as  $N/2$  photons could be trapped inside a volume containing  $N$  atoms. This possibility, although contained in most of the theoretical treat-

ments of superradiant emission, of  $N$  two-level systems, has been explicitly mentioned only by Freedhoff et al. [5], who introduced for the first time the word "subradiance", and by Stroud et al. [6]. However, none of these authors derived realistic conditions which would allow the observation of the phenomenon. It is at least clear that it does not occur when all the  $N$  atoms are initially excited, as it is the case in all superradiance experiments which have been performed up to now.

When looking for realistic conditions for an eventual observation of radiative trapping of a large number of photons, the case of many-level atoms opens new possibilities. In this note, we study a specific case of three-level atoms, for which both constructive and destructive interatomic interferences generally play a role in the evolution of the system, even when all the atoms are initially excited. In other words, both superradiance and subradiance phenomena are generally present in the cooperative spontaneous evolution of a collection of  $N$  such atoms. Any theoretical model which accounts for superradiant emission should thus be able to describe subradiance; this will be shown in the particular case of the semi-classical model. However this model does not provide us with general laws concerning subradiance and it does not exhibit clearly the physical origin of the phenomenon. As for a single pair of two-level atoms, subradiance is due to destructive interference and it is expected to depend upon the

symmetry properties of the atomic collective states. It is shown how the symmetry point of view can be introduced, in a clean way, in the study of cooperative spontaneous evolution of extended pencil-shaped samples. More specifically, symmetry considerations, based on group theory, allow then one to study quite generally the conditions in which subradiance could be observed. It is shown that one can find well-defined and realistic initial conditions giving rise to spontaneously created subradiant states, which are finally destroyed, but only by non-cooperative spontaneous emission.

In a first approach, the cooperative evolution of  $N$  three-level atoms such as represented in fig. 1 can be described in the semi-classical model. It is assumed here that all the atoms are initially excited, i.e. that they are either in state  $|1\rangle$  or in state  $|2\rangle$ , and that all initial atomic quantities are spatially homogeneous inside the active volume. Initial conditions for the evolution of the system are thus characterized by the values of the populations of the two upper levels and by the coherence between them. When computing the solution of the evolution equations, it appears that, for large values of  $t$ , the total population of the two upper levels reaches a constant (and spatially homogeneous) value which is generally different from zero; meanwhile, the coherence between these two upper levels tends to be complete. More precisely it is found that the limit value of the total population of the two upper levels is always zero (no subradiance) when the initial coherence is complete; this limit value can be as large as half the total population when the two initial populations are equal and when the initial coherence is zero <sup>†1</sup>. A numerical example cor-

<sup>†1</sup> This effect can also be understood as a quenching of the less probable transition by the other one [7].

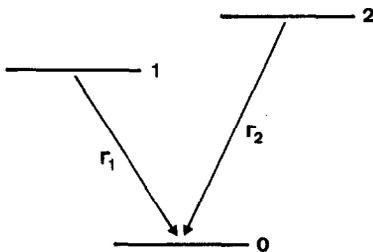


Fig. 1. Levels and transitions for a three-level atom; the polarizations or the frequencies of the two electric dipole transitions are assumed to be different.

responding to this latter situation, i.e. to a maximum subradiance effect, is shown in fig. 2.

The existence of this effect could have been predicted without computing the solutions of the evolution equations. Let us assume that the atoms are initially uncorrelated <sup>†2</sup>; this means that the initial atomic state can be represented by a factorizable density matrix

$$\rho(0) = \prod_{\alpha=1,N} \rho^{\alpha}(0), \tag{1}$$

$\rho^{\alpha}$  being a monatomic density matrix. Because of the spatial homogeneity,

$$\rho^{\alpha}(0) = \sum_{i,j} \rho_{ij}(0) |i\rangle_{\alpha\alpha} \langle j|, \tag{2}$$

<sup>†2</sup> The existence of initial interatomic correlations would not introduce any change in the solution of the Bloch-Maxwell equations.

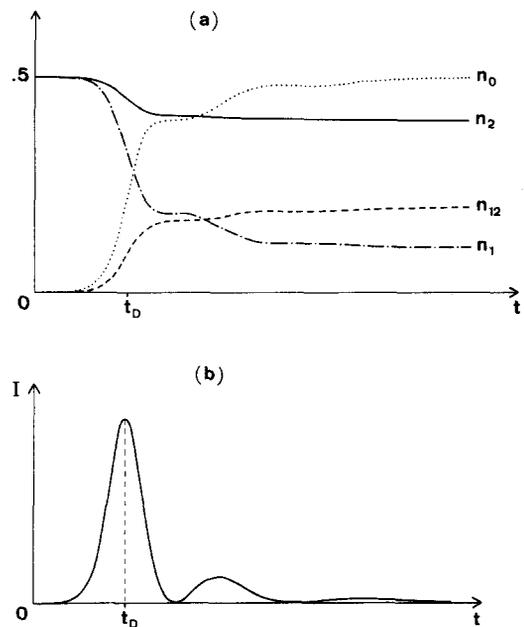


Fig. 2. Computed results of the semi-classical model with  $\Gamma_2/\Gamma_1 = 0.8$  and for initial conditions corresponding to  $n_1(0) = n_2(0) = 0.5$  and  $n_{12}(0) = 0$ . Curves in (a) represent the time evolution of the populations and of the coherence between the two upper levels; curve in (b) represents the total intensity radiated on both transitions. The delay time  $t_d$  is about  $30 T_{SR1}$  (characteristic time for cooperative spontaneous emission on transition  $1 \rightarrow 0$ ).

the sum running over the pairs of atomic states. In the non-markovian semi-classical model, the density matrix remains factorizable (but not homogeneous during the evolution of the system and eq. (1) is valid at any time. Furthermore, the statistical mixing does not change during the evolution and one has:

$$\text{Tr}[\rho^\alpha(t)^2] = \sum_{i,j} \rho_{ij}^\alpha(t) \rho_{ji}^\alpha(t) = \sigma^2 = \text{const.} \quad (3)$$

At  $t = 0$ ,

$$\sigma^2 = \rho_{11}(0)^2 + \rho_{22}(0)^2 + 2|\rho_{12}(0)|^2, \quad (4)$$

and  $1/2 \leq \sigma^2 \leq 1$ . At the end of the evolution,

$$\rho_{10}^\alpha(\infty) = \rho_{20}^\alpha(\infty) = 0, \quad (5)$$

and therefore:

$$\sigma^2 = \rho_{00}^\alpha(\infty)^2 + \rho_{11}^\alpha(\infty)^2 + \rho_{22}^\alpha(\infty)^2 + 2|\rho_{12}^\alpha(\infty)|^2. \quad (6)$$

Unless the initial state is a pure state ( $\sigma^2 = 1$ ), it is obviously impossible to have  $\rho_{00}^\alpha(\infty) = 1$ , which explains that the computed value of the population of the upper levels cannot reach zero. Moreover this suggests that it might exist a connection between the initial statistical mixing, which is characterized by  $\sigma^2$ , and the limit value of the population of the upper levels. When analyzing the numerical results, it is found, in effect, that the proportion  $\tau$  of atoms remaining excited depends on  $\sigma^2$  only. It will be shown in the following that the exact relationship between  $\tau$  and  $\sigma^2$  can be obtained in a more general way.

Let us now consider the general hamiltonian of a collection of  $N$  atoms (supposed, at first, to be at rest) interacting via electric dipole interaction with the quantized electromagnetic field,

$$H = H_A + H_F + H_I; \quad (7)$$

the atomic hamiltonian  $H_A$  and the interaction hamiltonian  $H_I$  can be written as sums over the  $N$  atoms:

$$H_A = \sum_{\alpha=1,N} h_\alpha \quad (8)$$

$$H_I = - \sum_{\alpha=1,N} (E(z_\alpha) \cdot d_\alpha),$$

where  $h$  is the hamiltonian for one isolated atom,  $E(z)$  is the quantized electric field,  $z_\alpha$  and  $d_\alpha$  are respectively the position and the dipole operator of the atom  $\alpha$ . Let us assume that the volume occupied by the atoms

is pencil-shaped (with the  $z$  axis as symmetry axis); the Fresnel numbers corresponding to the two atomic wavelengths,  $\lambda_1$  and  $\lambda_2$ , are assumed to be of the order of unity. It is then assumed that the emitted light consists in two plane-wave packets travelling in both  $z$  directions, with frequencies centered on the two atomic frequencies. This approximation being, at first, considered as rigorously valid (the discussion of its validity will be done later on), it follows that  $E(z)$  depends on the  $z$  coordinate only. Therefore, all the atoms which are contained in a slice defined by  $z_0 \leq z_\alpha < z_0 + \Delta z$ , with  $\Delta z$  of the order of the wavelengths  $\lambda_1$  and  $\lambda_2$ , must be considered as undistinguishable<sup>†3</sup> for the field they can radiate. This implies that the spontaneous evolution of the whole atom-field system will conserve some local symmetry properties. Precisely, let us consider the whole volume as formed by successive thin slices  $S_k$ , whose thicknesses are of the order of  $\lambda_1$  and  $\lambda_2$ ; the total hamiltonian is then invariant under the permutations of the indices of the atoms of a given slice. It is to be emphasized that the different slices do not have independent evolution, but that they interact through the field they radiate. However, this interaction does not affect the local symmetry conservation.

An important remark concerning the role of the slices must be done now. One knows that a necessary condition for cooperative emission, in the case of two-level atoms is [8]

$$T_{SR} \ll T_{SP} \quad (9)$$

( $T_{SR}$  is the characteristic time for cooperative emission, whereas  $T_{SP}$  is the characteristic time for ordinary spontaneous emission). For pencil-shaped volumes, this condition can be written

$$1/N\Gamma\mu \ll 1/\Gamma, \quad (10)$$

where the form factor  $\mu$  is given by  $\mu = (3/8\pi)\lambda^2/a^2$  [9]. The number of atoms contained in a given slice (with thickness of the order of  $\lambda$ ) is of the order of  $n = N\lambda/L$  and condition (9) leads to

$$n \gg (8\pi/3) \mathcal{F} \quad (11)$$

<sup>†3</sup> This undistinguishability is of course not absolute. On the contrary, it is assumed that the wavefunctions of the different atoms do not overlap (see ref. [1]); this explains that the collective wavefunctions can have any symmetry type.

( $\mathcal{F} = a^2/L\lambda$  being the Fresnel number). The generalization of this result to the considered three-level atoms case is straight forward; the previous assumption concerning the Fresnel numbers associated to  $\lambda_1$  and  $\lambda_2$  implies that the number of atoms contained in a given slice  $S_k$  is  $n \gg 1$ . Therefore, cooperative effects appear only when large numbers of atoms are undistinguishable (in the plane-wave approximation) for the field they radiate.

The local symmetry conservation suggests the use of atomic basis states formed by products of collective states corresponding to the different slices  $S_k$ , each of these  $n$ -atom states having well-defined symmetry properties with respect to the permutation group  $\mathcal{S}_n$ . In effect these symmetry properties (which analogue, in the two-level atom case, is the cooperation number  $r$ ) will be conserved during the spontaneous evolution of the whole atom-field system. In the up to now published theoretical treatments of cooperative spontaneous emission of two-level atoms, only fully symmetric collective states have been explicitly written [e.g. 10]. This arbitrary simplification is justified as long as all of the atoms are initially excited, so that the initial collective state is fully symmetric. In the considered three-level atoms case, this justification does not hold any longer and one must also consider partially antisymmetric states. Precisely one must take into account all the collective states of the slices  $S_k$  which have well-defined symmetry properties with respect to  $\mathcal{S}_n$ . These states can be labeled according to the irreducible representations of  $\mathcal{S}_n$ ; these representations will be called in the following "symmetry types" and they are characterized by partitions of  $n$ , written  $\{l_1 l_2 l_3\}$  (with  $l_1 + l_2 + l_3 = n$  and  $l_1 \geq l_2 \geq l_3$ ). An example of an  $n$ -atom wavefunction with symmetry type  $\{l_1 l_2 l_3\}$  is obtained in constructing the product of:

(i)  $l_3$  fully antisymmetric three-atom wavefunctions, containing  $l_3$  atoms in each of the three monatomic states;

(ii)  $l_2 - l_3$  antisymmetric two-atom wavefunctions involving pairs of monatomic states and containing, in all,  $2(l_2 - l_3)$  atoms;

(iii)  $l_1 - l_2$  monatomic states.

It is clear that such a wavefunction <sup>#4</sup> vanishes when the same monatomic state appears more than once in a given antisymmetric part. Therefore, the maximum number of atoms in a given state is not equal

to  $n$  (unless  $l_2 = l_3 = 0$ , i.e. the state is fully symmetric). In particular, the number of deexcited atoms contained in a state with symmetry type  $\{l_1 l_2 l_3\}$  is necessarily smaller than or equal to  $l_2 + l_3$ . Since, as already mentioned, the symmetry types of the different slices are conserved during the evolution, the knowledge of these symmetry types at  $t = 0$  fixes a minimum value for the proportion  $\tau$  of atoms which remain excited. For instance  $\tau$  can be equal to zero (no subradiance) if the initial symmetry types are  $\{n 0 0\}$  (fully symmetric state); the opposite case corresponds to the "most antisymmetric" symmetry type,  $\{n/3 n/3 n/3\}$ , and one has then  $\tau \geq 2/3$ . More generally,  $\tau$  is necessarily larger than or equal to

$$\tau_{\min} = \sum_k \sum_{\{l_1 l_2 l_3\}} f_k(\{l_1 l_2 l_3\})(l_2 + l_3)/n, \quad (12)$$

where the first sum runs over the indices of slices and the second one runs over all possible symmetry types;  $f_k(\{l_1 l_2 l_3\})$  is the partial trace of the initial atomic density matrix corresponding to the slice  $S_k$  over the space of the irreducible representation  $\{l_1 l_2 l_3\}$ .

In this treatment, the importance of subradiance depends upon the (conserved) local symmetry properties of the atomic density matrix. This is easily understood in terms of interatomic interference, when remembering the case of a pair of two-level atoms. In the final subradiance state of the system, the photons are trapped inside given slices, because of destructive interatomic interference (the different slices do not radiate any longer and can therefore be considered as independant. Moreover, in analogy with the two-atom case, the partial antisymmetry of the states determines the importance of the destructive interferences, and therefore, the minimum value for the proportion  $\tau$  of atoms remaining excited. It is shown hereafter that, in an important particular situation,  $\tau_{\min}$  depends upon the initial statistical mixing only, like it does in the semi-classical model. Let us assume that the initial atomic state has the following properties: the  $N$  atoms are either in state  $|1\rangle$  or in state  $|2\rangle$ , they are uncorrelated and the atomic

<sup>#4</sup> The use of such a wavefunction for representing the symmetry type  $\{l_1 l_2 l_3\}$  is slightly incorrect. One should, according to the Young tableaux theory, modify it by symmetrizing operators. However the present form is much simpler and leads hereafter to rigorously exact results.

properties are spatially homogeneous. The initial atomic matrix is thus represented by eqs. (1) and (2). For such uncorrelated atoms, the symmetry properties are in fact determined by the statistical properties. The possible symmetry types are  $\{l_1 l_2 0\}$ <sup>‡5</sup> and a partial trace  $f_k(\{l_1 l_2 0\})$  represents the probability of finding the slice  $S_k$  in a state of symmetry type  $\{l_1 l_2 0\}$ . A tedious calculation, based on Young tableaux theory, allows one to evaluate these probabilities and one finds that, for a given value of  $\sigma^2$  (defined by eq. (4)), the most probable symmetry type is such that

$$\begin{aligned} l_1 &= \frac{1}{2}n(1 + \sqrt{2\sigma^2 - 1}), \\ l_2 &= \frac{1}{2}n(1 - \sqrt{2\sigma^2 - 1}), \end{aligned} \quad (12)$$

moreover, as  $n \gg 1$  provided that eq. (9) holds, the corresponding probability is found to be close to one, so that

$$\tau_{\min} = \frac{1}{2}(1 - \sqrt{2\sigma^2 - 1}) \quad (14)$$

The way in which the system, starting from given initial conditions, reaches finally some subradiant state and the exact value of the proportion  $\tau$  of atoms which remain excited cannot be quantitatively studied in this general model. It is however clear that the initial state cannot be fully antisymmetric and that constructive interatomic interference must also appear. The partial deexcitation of the system which creates the subradiant state is achieved by cooperative emission, and, therefore, within a time-scale of the order of  $T_{\text{SR}}$ . In addition, the total number of emitted photons tends to be as large as possible so that  $\tau$  will always be equal to  $\tau_{\min}$ . These qualitative remarks are quantitatively confirmed by the results of the semiclassical model. Fig. 2 shows, in a particular example, how subradiance appears after the rapid emission of a superradiant pulse; in all the computations we have performed, the value of  $\tau$  is equal to the value of  $\tau_{\min}$  given by eq. (14), to quite a good approximation.

In the preceding treatment, simple symmetry considerations have allowed us to understand the subradiance phenomenon and to predict a minimum value for the proportion  $\tau$  of atoms remaining excited, which, in the important particular case of initially uncorrelat-

ed atoms, depends simply on the initial statistical mixing. It is to be emphasized here that these results are based on the local symmetry conservation<sup>‡6</sup> and that they are fully valid only for atoms at rest, in the plane-wave approximation and provided that the threshold condition (9) is fulfilled.

The approximations must be discussed now. First, the local symmetry conservation is not actually perfect, even in the plane-wave approximation. The components along the  $z$  axis of the distances between the atoms of a given slice are not zero but only smaller than the wavelengths; therefore, the interatomic interference cannot be rigorously fully destructive. In addition, it is obvious that the plane-wave approximation fails both at the beginning and at the end of the emission. When the field emitted in the  $z$  direction is small, the emission in the other directions can be preponderant and the local symmetry conservation does not hold, even approximately. In other words, emission processes which do not conserve the symmetry types of the slices (which are similar to the  $r$ -non conserving processes [12] in the two-level atoms case) can become important. Concerning the beginning of the emission, this problem has already been discussed, for two-level atoms [12]; it is expected – and observed – that the plane-wave approximation describes correctly the main features of the cooperative emission. Concerning the end of the emission, a direct consequence of the failure of the symmetry conservation is that the subradiant states will be destroyed by ordinary spontaneous emission in all but the  $z$  directions.

Concerning the influence of the motion of the atoms, several remarks can be made. First it is possible to introduce the Doppler broadening in the semiclassical model [8]. It appears, provided that the usual threshold condition [8]

$$T_2^* \ll T_{\text{SR}} \quad (15)$$

( $T_2^*$  being the Doppler characteristic dephasing time), holds for both atomic transitions, that the superradiant emission is not seriously modified; in addition

<sup>‡5</sup> One cannot have  $l_3 \neq 0$  because two monatomic states only are occupied.

<sup>‡6</sup> The hamiltonian defined by eqs. (7) and (8) contains the dipole-dipole interaction and the counter-rotating terms. However the frequency shifts due to the dipole-dipole interaction are able to destroy the local symmetry conservation. It is assumed here, as it is usually done for extended pencil-shaped samples, that this interaction is negligible. On this subject see, for instance ref. [11].

the final subradiant state is exactly the same as in the lack of Doppler broadening. However, in this treatment, the atoms are assumed to be at rest. As far as superradiant emission is concerned, this is approximately correct, because of condition (15). When dealing with subradiance, i.e. when considering an unrestricted time-scale, one has to be more careful. In fact the atoms do not remain confined in a given slice during periods of time much larger than  $T_2^*$  and, then, the local symmetry conservation loses its significance. Therefore the system actually reaches a subradiant state but, during its further evolution, it will radiate even in the  $z$  directions; however, as the motion of the atoms does not change the population inversion on the two transitions, such an emission will not be cooperative. Finally, in a realistic situation, the subradiant states are destroyed in a time which is of the order of the spontaneous lifetimes of the two upper levels. This however does not prevent one from observing the subradiant phenomenon, since the subradiant state is created in a time-scale which is of the order of the characteristic times for cooperative emission, i.e. much smaller than the spontaneous lifetimes. A more general and detailed study of subradiance will be given elsewhere [13].

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