

## Cooperative Phenomena in Resonant Electromagnetic Propagation

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Phenomena of coherent resonant propagation can be considered as resulting from the cooperative interaction of a certain number of excited two-level systems. It is shown that these phenomena can be characterized by a specific "maximum cooperation number" and by the associated "cooperation time." These are defined for the superradiant state, but their meaning and usefulness can be extended to other situations. The alternative description of superradiance as a spontaneous or as a stimulated effect is also discussed and it is shown that with the help of the new concepts, the Dicke quantum perturbative treatment can be reconciled with the semiclassical theories.

### I. INTRODUCTION

The problem of the electromagnetic emission from an assembly of excited nonoverlapping two-level atoms interacting through the common radiation field has been the subject of active research for the last 25 years. A sufficiently general quantum-mechanical solution is still lacking, but two approximate approaches have been very successful in describing particular situations.

One of these is that of Dicke,<sup>1</sup> who introduced the concept of cooperation in spontaneous emission processes (superradiance). In the simplest description of superradiance, Dicke takes, as the initial state, a collective state which is a simultaneous eigenstate of the total atomic-energy operator and of the total pseudoangular momentum (each two-level atom being described by Pauli operators). The *cooperation number* is equal to the eigenvalue of the pseudoangular momentum, and is a measure of the strength of atomic cooperation. We note that in such a state, which we call a *Dicke state*, the expectation value of the electric dipole operator is zero. Hence no macroscopic source is available for the electromagnetic (em) field, and the only radiative contributions arise from spontaneous emission.

On the other hand, most treatments of radiative processes in quantum optics are done in the *semiclassical* approximation.<sup>2-4</sup> In this approach, the atoms do not have quantum correlations, i. e., the total atomic state is the direct product of the individual atomic states. These are taken as superpositions of the two energy eigenstates, displaying a nonzero electric dipole, so that an over-all macroscopic polarization is available that acts as a source for the classical em field. These states

shall be designated Bloch states, by analogy with the magnetic resonance case.<sup>2</sup>

In some instances it may appear that these two approximate approaches lead to different results. One objective of the present paper is to show how the Dicke quantum perturbative treatment can be reconciled with the semiclassical treatment. To this effect, two new concepts are introduced: the *maximum cooperation number* and the *cooperation time*. The maximum cooperation number  $N_c$  is an upper bound to the number of centers which can be prepared to the superradiant state or can cooperate to superradiant emission. It will turn out that this number plays a critical role in resonant propagation phenomena at optical frequencies ( $\pi$ ,  $2\pi$ , and echo pulses). The cooperation time is the shortest possible decay time for a superradiant state which emits initially into the vacuum of photons.

A detailed treatment of the connections between Dicke and Bloch states falls beyond the scope of the present paper, and is given elsewhere.<sup>5</sup>

### II. NEW CONCEPTS DEFINED WITHIN THE PERTURBATIVE QUANTUM APPROACH

Let us consider the coherent interaction of a radiation field with an assembly of otherwise independent  $N$  two-level systems. The atoms are distributed with a density  $\rho$  over a region of space large compared to the wavelength  $\lambda$ , so that retardation effects are important. In this and in Sec. III, it is assumed that the atoms have all been prepared in a properly phased superradiant state, at some initial instant of time, and in the vacuum of photons. The phase is that of a plane wave of direction  $\vec{k}$ . In this section we assume that this initial state is a superradiant Dicke state, i. e., an eigenstate of the unperturbed atomic energy, having an energy

quantum number  $M \approx 0$ , and the maximum total angular momentum  $\frac{1}{2}N$ . This initial condition is the most convenient one for the introduction of the new concepts of cooperation time and maximum cooperation number. It may however be very difficult to prepare a sample in this initial state, as will be discussed in Sec. IV.

Using first-order-perturbation theory, the emission intensity in the direction  $\vec{k}'$  is given by<sup>1</sup>

$$I(\vec{k}') = I_0(\vec{k}') \left( \frac{1}{4} N \right) (1 + N |\langle e^{i(\vec{k}-\vec{k}') \cdot \vec{x}} \rangle|^2). \quad (1)$$

Here  $I_0(\vec{k}')$  is the spontaneous intensity radiated by a single isolated atom in the direction  $\vec{k}'$ , and the angular brackets denote an average over the atomic positions  $\vec{x}$ . Assuming that the sample is a rod of cross section  $A$ , and that  $\vec{k}$  is chosen along the rod axis, the interference exponential in Eq. (1) causes the emission to be mainly confined to a cone of aperture  $\Delta\Omega \approx 4\pi\lambda^2/A$  around the direction  $\vec{k}$ .<sup>6</sup> Here  $\lambda$  is the wavelength in the medium. The emission rate is enhanced by the factor

$$\frac{1}{4}(1 + N |\langle e^{i(\vec{k}-\vec{k}') \cdot \vec{x}} \rangle|^2),$$

which is of the order of  $\frac{1}{4}N$  within this cone. Denoting the spontaneous emission rate of an isolated atom over the full solid angle by  $\gamma$ , the spontaneous rate of an isolated atom over the reduced angle is  $\gamma\Delta\Omega/4\pi$ , and the enhanced rate per atom is  $\frac{1}{4}N$  times larger, i. e.,

$$\gamma_D = \frac{1}{4}N\gamma\lambda^2/A. \quad (2)$$

This Dicke rate corresponds to the linewidth of the superradiantly emitted photons, i. e., to the reciprocal of their decay time. In this first-order-perturbation treatment it has been assumed that  $N$  is the total number of two-level systems within the sample. But in the optical case,  $N$  can be very large; and the uncritical use of Eq. (2) with very large  $N$  can lead to a clearly unrealistic decay rate (e. g., a decay rate much faster than the carrier frequency of the radiation).<sup>7</sup> This difficulty can be traced back to assumptions which are made in the derivation of Eq. (1). In particular, this equation assumes that the enhanced radiation rate  $\gamma_D$  that it predicts is smaller than both  $\omega$  and  $c/l$ , where  $\omega$  is the optical frequency,  $l$  is the sample length, and  $c$  the light velocity in the medium. The first of these conditions ( $\omega > \gamma_D$ ) is related to the fact that the first-order-perturbation result is obtained by averaging over many optical periods. The second condition ( $c/l > \gamma_D$ ) is due to the fact that Eq. (1) takes retardation into account as if the emitted radiation were an undamped plane wave. For optical frequencies, it is usually this second condition which first breaks down as  $N$  becomes too large, or, as will be seen shortly, as the atomic density becomes too small for a fixed  $N$  value. A model calculation with the same initial condition as here, but re-

stricted to two atoms, is developed in the Appendix. In this case,  $l$  is simply the distance between the two atoms; it is found that if  $l$  is smaller than  $\sim c/2\gamma$  the two atoms decay essentially together with a rate of  $2\gamma$  (which is twice the normal decay rate, since in this case there is initially a single excitation in the system). On the other hand, if  $l$  is larger than  $\sim c/2\gamma$  the two atoms decay independently, and the total decay rate is  $\gamma$ . It is clear from the analysis of the Appendix that the enhanced decay starts after the radiation from either of the atoms has reached the other atom. In other words, the effect of the distance on the decay rate is due to the combined action of propagation and damping. Similarly for the case of  $N$  atoms, one expects that the enhanced decay rate  $\gamma_D$  of Eq. (2) will be realistic only if the entire sample is shorter than  $c/\gamma_D$  so that all atoms in the sample are exposed to the radiation from all the others before the decay process is completed. The question we ask then is "What is the appropriate decay rate  $\gamma_c$  for a sample of size larger than  $c/\gamma_D$ ?"

The answer can be obtained via a self-consistency argument. The number of atoms which can be covered by the radiation of one of them during the decay time  $1/\gamma_c$  is given by

$$N_c = (c/\gamma_c) A \rho. \quad (3a)$$

These atoms superradiate with a rate given by first-order-perturbation theory, i. e., by the Dicke formula (2)

$$\gamma_c = \frac{1}{4}N_c\gamma\lambda^2/A. \quad (3b)$$

These two relations can be solved self-consistently for  $\gamma_c$  and  $N_c$ :

$$\gamma_c \equiv 1/\tau_c = \frac{1}{2}(c\rho\gamma\lambda^2)^{1/2}$$

and

$$N_c = (2A/\lambda) (c\rho/\lambda)^{1/2}, \quad (4)$$

where  $\tau_c$  is defined as the *cooperation time*.  $N_c$  is the *maximum cooperation number*, i. e., the maximum number of atoms that can cooperate to superradiant emission in a particular experimental situation. Assuming that a sample could be prepared in the given initial condition, and with  $N > N_c$ , all  $N$  atoms emit of course, but only  $N_c$  of them cooperate. This means that the emission rate is not given by (2) but by (3b). The *maximum cooperation length*  $c\tau_c$  is independent of the sample geometry and depends only on the transition parameters and on the density.

Order-of-magnitude values of  $c\tau_c$  are typically  $10^4$  cm in NMR (at 1 MHz), 10 cm in ESR (at 10 GHz), and 0.1 cm for the 6943-Å transition in ruby with 0.05% Cr<sup>3+</sup> concentration. The limited cooperation length becomes, therefore, of prime impor-

tance in optical experiments.

### III. CONNECTION WITH SEMICLASSICAL APPROACH

The self-consistent argument of Sec. II was based on a perturbative quantum treatment in line with Dicke's considerations. However, it is expected that a semiclassical treatment based on Maxwell equations for the field, and on Bloch equations for the atoms, leads to identical concepts of limited cooperation. In this case, it is assumed that at the initial time the atoms are in a properly phased superradiant Bloch state. The phase is that of a plane wave, and the Bloch vector is initially tipped by an angle  $\varphi = \frac{1}{2}\pi$  (the angle is measured from the ground state,  $\varphi = 0$ ).

The atoms which have been prepared in a superradiant Bloch state act as classical dipole sources for Maxwell equations. For a very long sample, the initial condition is uniform in the propagation direction. We can predict, without making any calculation, that the maximum field which will eventually be built in the sample volume corresponds to the release of an energy  $\frac{1}{2}\hbar\omega$  per atom, i. e., to an energy density<sup>8</sup>  $\epsilon E^2 = \frac{1}{2}\hbar\omega\rho$ , where  $\epsilon$  is the optical dielectric constant of the host material. On the other hand, the Rabi precession by an angle  $\frac{1}{2}\pi$  which is induced by this field takes a time  $\tau' \simeq \frac{1}{2}\pi \times (\hbar/2\mathcal{O}E)$ , where  $\mathcal{O}$  is the transition dipole moment. The spontaneous rate  $\gamma$  is proportional to  $\mathcal{O}^2$  (see Ref. 9). Combining the two relations one obtains, within a numerical factor of order 1,  $\tau' \simeq \tau_c$ , and therefore we recover the cooperation time with the same meaning as in the quantum treatment. In this case there is no need to make a self-consistency reasoning, since the energy balance equation  $\epsilon E^2 = \frac{1}{2}\hbar\omega\rho$  has both propagation and damping automatically built in. For these considerations to hold, the cooperation length  $l_c = c\tau_c$  must be shorter than the sample length  $l$ . If  $l_c > l$ , the photons leak out of the sample during the emission process, and the available energy density is reduced by the ratio  $l/l_c$ . In such a case the above argument yields the original result of Dicke [Eq. (2)], i. e., atoms in the sample contribute together to superradiant emission.

As opposed to the full quantum treatment, this semiclassical argument is, of course, very easily formalized. Within the slowly varying envelope approximation the problem is described by the following equation<sup>10</sup>:

$$\frac{\partial^2 \varphi}{\partial t^2} + c \frac{\partial^2 \varphi}{\partial t \partial z} = -\omega_c^2 \sin \varphi, \quad (5)$$

where  $t$  is the time and  $z$  the spatial coordinate in the direction of propagation. The frequency  $\omega_c$  is given by

$$\omega_c^2 = \frac{\omega\rho\mathcal{O}^2}{\hbar\epsilon} = \frac{1}{\pi\tau_c^2}, \quad (6)$$

in which  $\tau_c$  has the value given by (4). The envelope of the radiated field is

$$E = \frac{\hbar}{2\mu} \frac{\partial \varphi}{\partial t}. \quad (7)$$

For an infinitely long sample,  $\varphi$  is a function of  $t$  only, so that the solution to (5) is immediately given in terms of elliptic functions.<sup>11</sup> The em energy is periodically emitted and reabsorbed, and the quarter-period of the process is  $\tau' = (1/\omega_c)F(\frac{1}{2}\sqrt{2}, \frac{1}{2}\pi) \simeq 3.3\tau_c$ , where  $F$  is the elliptic integral of the first kind.

### IV. FURTHER MEANING OF THESE CONCEPTS

In Secs. I–III it was implicitly assumed that a sample can be initially prepared to a superradiant state over an arbitrary length. But, in fact, the cooperation length  $l_c = c\tau_c$  also puts an upper limit to the sample length which can be prepared in the superradiant state by the action of a classical field. For instance, we could plan on preparing the sample by a  $90^\circ$  pulse of very short duration. Since the energy of a  $90^\circ$  pulse is inversely proportional to its duration, the pulse could supposedly be made so short that it loses essentially no energy upon traversing the sample. In these most favorable conditions, the length of sample which can be excited *at one time* to a state that approaches the  $90^\circ$  state turns out to be limited to  $l_c$  as will be shown presently.

The solution to this problem is obtained using Eq. (5) which applies for times shorter than the homogeneous relaxation time of the medium. We let the exciting pulse impinge on the entrance face  $z = 0$  at time  $t = 0$ . By transforming the coordinates to

$$\xi = \omega_c(t - z/c) \quad (8a)$$

and

$$\zeta = \omega_c z/c, \quad (8b)$$

Eq. (5) becomes

$$\frac{\partial^2 \varphi}{\partial \xi \partial \zeta} = -\sin \varphi. \quad (9)$$

Approximating the exciting pulse by a  $\delta$  function we obtain the boundary condition  $\varphi = \frac{1}{2}\pi$  for  $\xi = 0$ . Since we have considered only one direction of propagation, and within the slowly varying envelope approximation, the atoms situated exactly at the entrance face  $z = 0$  will never radiate. Therefore the second boundary condition is  $\varphi = \frac{1}{2}\pi$  at  $\zeta = 0$ . This somewhat unphysical boundary condition is necessary in order to avoid having to consider the problem of the reflected wave,<sup>12</sup> but, except for the ab-

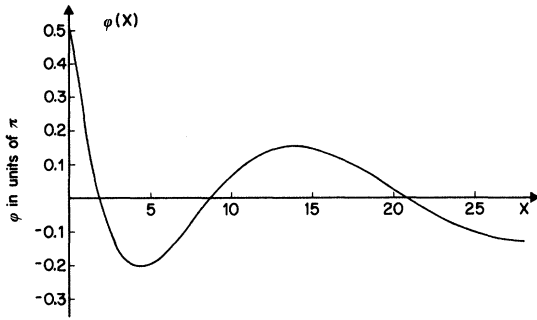


FIG. 1. The function  $\varphi(x)$ .

sence of rounding off at the edges, this should not affect the main result.

With this symmetric boundary condition, it turns out that the solution to Eq. (9) is of the form  $\varphi = \varphi(x)$ , where  $x = \xi\zeta$ . Indeed, the initial condition is  $\varphi = \frac{1}{2}\pi$  for  $x=0$  and the equation can be written in a form which depends on  $x$  only:

$$x \frac{d^2\varphi}{dx^2} + \frac{d\varphi}{dx} = -\sin\varphi. \quad (10)$$

Imposing that the second derivative  $d^2\varphi/dx^2$  be finite at  $x=0$ , we obtain a second initial condition  $d\varphi/dx=1$ . The solution is plotted in Fig. 1.<sup>13</sup> We note that the sample excitation, at a given time  $t$  and as a function of position  $z$ , is obtained by writing  $x = \omega_c^2(t - z/c)z/c$  in the solution  $\varphi(x)$ . The new function which is thus obtained is plotted versus  $Z = \omega_c z/c$  in Fig. 2 for several values of time  $T = \omega_c t$ . This plot immediately shows that, up to a depth of the order of the cooperation length, the sample can be excited to superradiance, after which this state decays in a time which is of the order of the cooperation time. This justifies our further interpretation of  $l_c$ .

We also note that a very different manner of preparing a sample to superradiance can be conceived, as originally pointed out by Dicke.<sup>1</sup> This is to invert the sample completely, and to let it decay by *spontaneous emission*. There is no theoretical limit to the size of sample that can be inverted, and therefore it is worthwhile knowing how a sample would decay if its length  $l$  were much greater than  $l_c$ . No theoretical answer to this complicated question is yet available, but an educated guess can be made on the basis of the present considerations. We expect that the sample will exhibit a spiking emission, dividing into random superradiant regions of average length  $l_c$ , and with no phase correlation between different regions. That  $\tau_c$  should play an important role in this case can be understood in very simple terms. A large sample has linear dimensions larger than  $c\tau_c$ , and up to time  $\tau_c$ , the emitted photons can be considered as com-

pletely contained within the sample. The number of radiation modes of one polarization contained in the sample volume  $V$ , is  $p = (4\pi V/\lambda^3) \times (\Delta\omega/\omega)$ . At  $t = \tau_c$  the spectral width is still equal to the observational spectral width, which is  $\Delta\omega \approx 2/\tau_c$ . It is then easy to verify that at  $t = \tau_c$  the number of spontaneously emitted photons  $\rho V \gamma \tau_c$  is equal to the number of modes  $p$  that can be supported by the sample volume. Up to  $\tau_c$  the rate is the spontaneous single atom rate  $\gamma$ , but as soon as there is one photon per radiation mode, stimulated effects become important and the nature of the emission changes.<sup>14</sup> Moreover, since the initial photons were emitted spontaneously, there is no phase correlation between them, and therefore we expect no phase correlation between the various superradiant regions.

#### V. ROLE OF COOPERATION TIME IN RESONANT em PROPAGATION

As several semiclassical calculations of resonant em propagation are already available, it is of interest to see how the concepts of limited cooperation can be recovered from their results. In particular, the known  $\pi$  and  $2\pi$  solutions<sup>10,15,16</sup> must all have pulse durations which are smaller than, or equal to, the cooperation time, otherwise the pulse coherence could not be maintained during propagation.

In order to carry out our task, the self-consistent argument of Sec. II [Eqs. (3)] has to be refined to account correctly for inhomogeneous broadening, distributed nonresonant losses, or finite length of the excited section of the sample. Equation (3b), which describes the result of first-order-perturbation theory, is not affected by these factors, but the number of cooperating atoms, given by Eq. (3a), may have to be modified. The effect of inhomogeneous broadening is to reduce the effective density of two-level systems from  $\rho$  to  $\rho T_2^*/\tau$ , where  $T_2^*$  is the inverse of the inhomogeneous linewidth and  $\tau$  the exciting pulse width, and where it is assumed that  $\tau > T_2^*$ . This is due to the finite bandwidth  $\tau^{-1}$  of the fully coherent exciting pulse. A nonresonant loss introduced into Maxwell equations by a conductivity

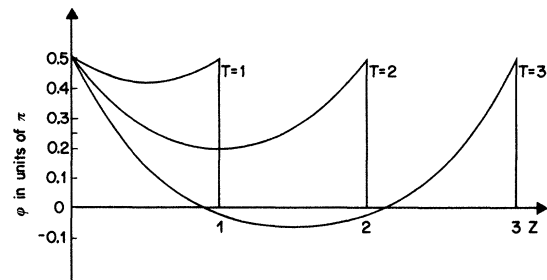


FIG. 2. Sample excited by a short  $\frac{1}{2}\pi$  pulse: Tipping angle of the Bloch vector as a function of  $Z = \omega_c(z/c)$  for various times  $T = \omega_c t$ .

TABLE I. Modifications to the right-hand side of Eq. (3a). The symbols are defined in the text.

The effect of:	is to replace the quantity:	by the following quantity, provided the latter is smaller than the former
Inhomogeneous broadening $T_2$	$\rho$ (density)	$\rho T_2^*/\tau$
Distributed nonresonant losses $\sigma$	$c/\gamma_c$ (cooperation length)	$l_a = 2c\epsilon/\sigma$
Finite excited length of sample $l_p$	$c/\gamma_c$ (cooperation length)	$l_p = V\tau$

$\sigma$  imposes an upper limit to the length which can be set in cooperation. This limit is the absorption length for the field  $l_a = 2c\epsilon/\sigma$ . Finally, a finite pulse length  $l_p = V\tau$  (here  $V$  is the pulse propagation velocity) also puts an upper limit to this length, since the material system is not excited beyond  $l_p$ . For clarity, these considerations have been summarized in Table I.

We now calculate the appropriate cooperation time for the following cases: long  $2\pi$  pulses ( $\tau \gg T_2^*$ ), short  $2\pi$  pulses ( $\tau \ll T_2^*$ ),  $\pi$  pulses in a system with a narrow homogeneous line and large nonresonant losses ( $\tau > 2c\epsilon/\sigma$ ),  $\pi$  pulses in a system with a narrow homogeneous line and small nonresonant losses ( $\tau < 2c\epsilon/\sigma$ ). The results have been summarized in Table II. For each one of these cases the following approach is taken: First a guess is made as to what is the appropriate limit length which has to be used in Eq. (3a) (second column of Table II). Then  $N_c$  is calculated (third column), and the appropriate pulse velocity  $V$  or pulse duration  $\tau$  is taken from the corresponding semiclassical theory (fourth column).<sup>10,15</sup> Using these data,  $\tau_c = 1/\gamma_c$  is obtained from Eq. (3b), and is compared to the actual pulse length  $\tau$  (fifth column). Finally, the validity of the initial assumption on the limit length is checked

(sixth column).

The fifth column of Table II immediately shows that, besides a factor which results from the definition of the various quantities and which is of course irrelevant, propagating  $\pi$  and  $2\pi$  pulses have a duration which is not longer than the cooperation time. We note that there is an additional factor of 2 which enters the inequality in the case of  $\pi$  pulses. This can be ascribed to the fact that in the  $\pi$ -pulse case there is only superradiant emission, whereas in the  $2\pi$ -pulse case there is superradiant emission for half of the pulse duration and absorption for the other half. Moreover, we note that long  $2\pi$  pulses, or long  $\pi$  pulses (large nonresonant losses), have their duration limited by  $\tau_c$ . This corresponds to the fact that, in these cases, the em energy density at the peak of the pulse is considerably smaller than the energy density in the two-level atoms (as shown for  $2\pi$  pulses in Ref. 17), so that the emission takes place essentially into the photon vacuum. On the other hand, the pulse duration is shorter than  $\tau_c$  for energetic  $\pi$  and  $2\pi$  pulses; there is no contradiction in this result, since the cooperation time was defined for a superradiant state starting to emit in the photon vacuum. If a large field is already present, the emission time is further reduced.

TABLE II. The limited-cooperation concept in some semiclassical calculations.

Case of interest	Limit length in Eq. (3)	Cooperation number $N_c$	Result of the semiclassical theory (Refs. 10 and 15)	Value of the cooperation time $\tau_c$ compared to $\tau$	Check on the assumed limit length
Long $2\pi$ pulses $\tau \gg T_2^*$	$l_p = V\tau$	$\rho A l_p T_2^*/\tau$	$V \approx 2/\alpha\tau$ with $\alpha = 2\omega\rho\mathcal{P}^2 T_2^*/\epsilon c\hbar^a$	$\tau_c \approx \frac{2\alpha\tau}{\rho T_2^* \gamma \lambda^2} = \frac{\tau}{\pi}$	$V\tau < c\tau_c$ since $V \ll c$
Short $2\pi$ pulses $\tau \ll T_2^*$	$l_p = V\tau$	$\rho A l_p$	$V = cT^2/(T^2 + \tau^2)$ with $T^2 = \epsilon\hbar/\rho\omega\mathcal{P}^2{}^a$	$\tau_c = \frac{\tau}{\pi} \frac{T^2 + \tau^2}{\tau^2} > \frac{\tau}{\pi}$	$l_p = \frac{cT^2\tau}{T^2 + \tau^2} \lesssim$ $l_c = \frac{c\tau}{\pi} \cdot \frac{T^2 + \tau^2}{\tau^2}$
$\pi$ pulses: large losses $\tau > 2c\epsilon/\sigma$	$l_a = 2c\epsilon/\sigma$	$\rho A l_a$	$\tau = \frac{\pi\sigma}{\gamma\rho\lambda^2 c\epsilon}$	$\tau_c = \frac{2\sigma}{\gamma\rho\lambda^2 c\epsilon} = \frac{2\tau}{\pi}$	$l_a < l_p \approx l_c$ since $\frac{2\epsilon}{\sigma} < \tau$
$\pi$ pulses: small losses $\tau < 2c\epsilon/\sigma$	$l_p = V\tau$	$\rho A l_p$	$\tau = \frac{\pi\sigma}{\gamma\rho\lambda^2 c\epsilon}$	$\tau_c = \frac{4\epsilon}{\pi\sigma} > \frac{2\tau}{\pi}$	$l_p \lesssim l_c < l_a$

<sup>a</sup>The value of  $\mathcal{P}^2$  is to be taken from Ref. 9.

## VI. CONCLUSIONS

The cooperation time has been defined as the emission time of a sample placed in the superradiant state and in the vacuum of photons. We have seen that this concept is intimately related to propagation effects. These propagation effects are automatically accounted for in treatments based on Maxwell wave equation, but are equally important in situations which have to be described quantum mechanically (for example, the emission from a Dicke state), in which case they are not always exhibited straightforwardly.

Besides this formal difference, we have seen that superradiance can be considered equally well as a spontaneous or as a stimulated effect. The two approaches can be reconciled by using the limited cooperation concept.

Of course, propagation effects are also important in situations described by other initial conditions, and therefore it should be expected that the cooperation time has a broader meaning than what is implied by its definition. In particular, we have considered the emission from a large sample which is initially completely in the upper state, and in the vacuum of photons. In this situation, the cooperation time defined by Eq. (4) turns out to be the time after which the atoms cease to emit individually and start to emit collectively.

We have also introduced the concept of a maximum cooperation number  $N_c$ . This number determines the sample size beyond which specific propagation effects play a major role, and is therefore of prime importance from the experimental point of view. For example, in the case of photon echoes, the purpose of the experiments is usually to determine specific properties of the two-level systems. These properties can be relaxation times, which affect the echo intensity,<sup>18</sup> or level degeneracy, which affects the polarization dependence of the echo.<sup>19</sup> It is usually not desired to observe propagation effects, which can only obscure the results. For instance the polarization dependence of the echo is strongly affected if the sample contains more than  $N_c$  resonant centers.<sup>20</sup> It is therefore important in designing actual echo experiments to take the maximum cooperation number into account.

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## APPENDIX A: EMISSION OF TWO ATOMS PLACED IN THE SUPERRADIANT STATE

We present here a formal treatment of the emission from two atoms which are initially placed in a

properly phased superradiant Dicke state in the presence of no photons. In the course of time, the system moves in the subspace spanned by the states  $|1\ 2; 0\rangle$ ,  $|2\ 1; 0\rangle$ , and  $|1\ 1; \vec{q}\rangle$ . The first two labels designate the state of the two atoms (1 for the ground state and 2 for the excited state). The third label indicates the state of the photon field (either no photons, or one photon in mode  $\vec{q}$ ). We consider photons of one polarization only, corresponding to the polarization of the two-level system transition. These are the only possible states, since the system contains only one excitation. In order to phase the wave functions as if they had been excited by a plane wave of wave vector  $\vec{k}$ , where  $\omega = ck$  is the resonant frequency, we multiply the wave function of an atom at position  $\vec{x}$  by  $e^{i/2\vec{k}\cdot\vec{x}}$  if the atom is in state 2, and by  $e^{-i/2\vec{k}\cdot\vec{x}}$  if the atom is in state 1. We note that a suitable orthonormal basis in the above subspace is given by

$$|\psi_1\rangle = (1/\sqrt{2})(|1\ 2; 0\rangle e^{i/2\vec{k}\cdot(\vec{x}_2-\vec{x}_1)} + |2\ 1; 0\rangle e^{i/2\vec{k}\cdot(\vec{x}_1-\vec{x}_2)}) , \quad (\text{A1})$$

$$|\psi_2\rangle = (1/\sqrt{2})(|1\ 2; 0\rangle e^{i/2\vec{k}\cdot(\vec{x}_2-\vec{x}_1)} - |2\ 1; 0\rangle e^{i/2\vec{k}\cdot(\vec{x}_1-\vec{x}_2)}) , \quad (\text{A2})$$

$$|\psi_3\rangle = |1\ 1; \vec{q}\rangle e^{-i/2\vec{k}\cdot(\vec{x}_2+\vec{x}_1)} , \quad (\text{A3})$$

where  $\vec{x}_1$  and  $\vec{x}_2$  are the positions of the two atoms.  $|\psi_1\rangle$  is the initial superradiant state.

The Hamiltonian of the system is

$$\begin{aligned} \mathcal{H}/\hbar = & \frac{1}{2}\omega(\sigma_1^z + \sigma_2^z) + \sum_{\vec{q}} \omega_{\vec{q}} a_{\vec{q}}^\dagger a_{\vec{q}} \\ & + \sum_{\vec{q}} [\kappa_{\vec{q}} (a_{\vec{q}}^\dagger \sigma_1^- e^{-i\vec{q}\cdot\vec{x}_1} + a_{\vec{q}}^\dagger \sigma_2^- e^{-i\vec{q}\cdot\vec{x}_2}) \\ & + \kappa_{\vec{q}}^* (a_{\vec{q}} \sigma_1^+ e^{i\vec{q}\cdot\vec{x}_1} + a_{\vec{q}} \sigma_2^+ e^{i\vec{q}\cdot\vec{x}_2})] , \end{aligned} \quad (\text{A4})$$

where  $\sigma_i^z$ ,  $\sigma_i^+$ ,  $\sigma_i^-$  are the Pauli-spin operators for atom  $i$ , and  $a_{\vec{q}}^\dagger$  and  $a_{\vec{q}}$  are the creation and annihilation operators for a photon in mode  $\vec{q}$ . The first term is the atomic energy, the second term is the field energy, and the third term is the interaction between the radiation field and the atoms. Since we are interested only in the damping, we shall neglect the dependence of the coupling constants  $\kappa_{\vec{q}}$  on  $\vec{q}$ , and replace them by  $\kappa = \kappa^*$  which is placed outside of the summation sign. Moreover, since we are making a calculation that pertains to a problem of superradiance, we expand the em field only in plane-wave modes having their propagation vectors parallel to the line joining the two atoms. In doing so, the result will be more directly applicable to superradiance problems involving a large number of atoms but with a preferential radiation direction which is

imposed by the preparation of the system. A physical situation to which our calculation directly applies is that of two atoms placed in a long waveguide which supports only one transverse mode. We denote by  $L$  the length of our one-dimensional cavity and assume reflecting boundary conditions so that  $q = n\pi/L$  (where  $n$  is a non-negative integer). The coupling constant  $\kappa$ , which in a three-dimensional problem contains a factor  $1/V^{1/2}$ , where  $V$  is the cavity volume, contains now a factor  $1/\lambda L^{1/2}$  instead. The time-dependent state of the system is written as

$$|\psi\rangle = b_1(t) |\psi_1\rangle + b_2(t) |\psi_2\rangle + \sum_q b_q(t) |\psi_q\rangle, \quad (\text{A5})$$

and the equation of motion of any of the coefficients  $b_j$  is simply

$$i \dot{b}_j = \langle \psi_j | \mathcal{H} | \psi \rangle. \quad (\text{A6})$$

Using (A1)–(A6) we obtain

$$i \dot{b}_1 = \frac{\kappa}{\sqrt{2}} \sum_q b_q (e^{i(q-k)x_2} + e^{i(q-k)x_1}), \quad (\text{A7})$$

$$i \dot{b}_2 = \frac{\kappa}{\sqrt{2}} \sum_q b_q (e^{i(q-k)x_2} - e^{i(q-k)x_1}), \quad (\text{A8})$$

$$i \dot{b}_q = (\omega_q - \omega) b_q + \frac{\kappa}{\sqrt{2}} [b_1 (e^{-i(q-k)x_2} + e^{-i(q-k)x_1}) + b_2 (e^{-i(q-k)x_2} - e^{-i(q-k)x_1})]. \quad (\text{A9})$$

We now take the Laplace transform of these equations with the initial condition  $b_1(0) = 1$ ,  $b_2(0) = b_q(0) = 0$ . The Laplace transform of  $b_i(t)$  is designated by  $B_i(p)$ . Taking the value of  $B_q$  from the transform of (A9), and substituting it in the transforms of (A7) and (A8), we obtain

$$\begin{aligned} pB_1 &= 1 - \kappa^2 B_1 \sum_q \frac{1 + \cos(q-k)l}{p + ic(q-k)} \\ &\quad + i\kappa^2 B_2 \sum_q \frac{\sin(q-k)l}{p + ic(q-k)}, \\ pB_2 &= -i\kappa^2 B_1 \sum_q \frac{\sin(q-k)l}{p + ic(q-k)} \end{aligned} \quad (\text{A10})$$

$$B_1(p) = \frac{p + \frac{1}{2}\gamma \coth(pl/c) - \frac{1}{2}\gamma \cosh(pl/c - pl/c)/\sinh(pl/c)}{[p + \frac{1}{2}\gamma \tanh(pl/2c)][p + \frac{1}{2}\gamma \coth(pl/2c)]}, \quad (\text{A12})$$

where

$$\gamma = 2\kappa^2 L/c \quad (\text{A13})$$

is the single atom decay rate.

It remains to take the inverse transform of (A12).

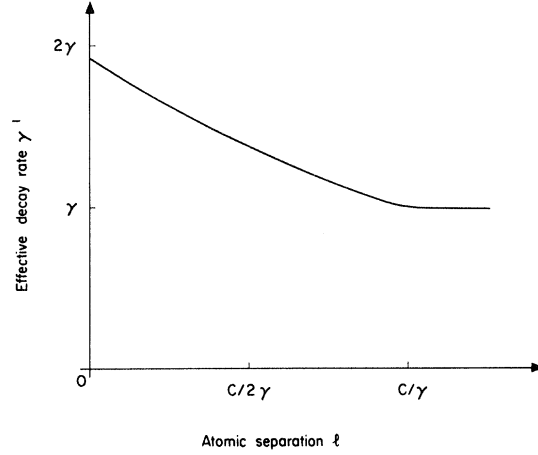


FIG. 3. Effective emission rate  $\gamma'$  of two atoms separated by a distance  $l$ . The atoms are initially placed in the superradiant state.  $\gamma'$  is defined as the inverse of the time at which the energy in the atomic system has decayed to  $1/e$  times the initial energy.

$$-\kappa^2 B_2 \sum_q \frac{1 - \cos(q-k)l}{p + ic(q-k)}, \quad (\text{A11})$$

where  $l = x_2 - x_1$ , is the distance between the two atoms. In the following we assume that  $l \geq 0$ . Since we intend to let  $L$  tend to infinity, we can assume without loss of generality that one of the resonant frequencies of the cavity coincides with the transition frequency  $\omega$ . Then  $q - k = \pi n/L$ , where  $n$  is an integer. Strictly speaking, the range of  $n$  is from  $-kL/\pi$  to  $+\infty$ . In these conditions the sums in (A10) and (A11) diverge. This is only a formal difficulty since in reality  $\kappa$  has a high-frequency cut-off. In fact, we may extend the range of  $n$  in the sums from  $-\infty$  to  $+\infty$  and take the principal value. The terms which are then added only affect the behavior on the very short time scale (on the scale of the inverse of the atomic frequency) which is not of interest here.

Performing the summation over modes as just stated, and solving for  $B_1$ , we obtain

It is rather easy to see that all the poles of  $B_1(p)$  are on the imaginary axis. If we let  $L$  tend to infinity, the linear density of poles increases proportionally to  $L$ , but their residues tend to zero like  $1/L$ . In this fashion, a branch cut is obtained, and the sum

over residues is replaced by an integral over this branch cut. The result of this integration is

$$b_1(t) = e^{-\gamma/2t} - \frac{1}{2}\gamma(t-l/c)e^{-(\gamma/2)(t-l/c)} U(t-l/c), \quad (\text{A14})$$

where  $U(t-l/c)$  is the unit step at  $t=l/c$ . [ $U(x) = 0$  for  $x < 0$ , and  $U(x) = 1$  for  $x > 0$ .] The same result can be obtained by letting  $L$  go to infinity before calculating the inversion integral. Taking the Bromwich integration contour at an abscissa where  $p$  has a finite positive real part, we have  $\lim \coth pL/c = \lim \tanh pL/2c = \lim \coth pL/2c = 1$ , and  $\lim \cosh(pL/c - pl/c) / \sinh pL/c = e^{-pl/c}$  on the integration line. The resulting function has a double pole at  $p = -\frac{1}{2}\gamma$ , and (A14) is obtained immediately. This expression gives the probability amplitude of finding the system in state  $|\psi_1\rangle$  at any time  $t > 0$ . The result is very interesting. For  $t \leq l/c$ , we see that the two atoms decay independently, giving  $|b_1|^2$

$= e^{-\gamma t}$ , i. e., a total decay rate which is the normal single excitation decay rate. For  $t > l/c$ , the decay rate is enhanced. In particular, for  $l=0$ , the initial rate is the superradiant rate  $2\gamma$ . In a similar fashion, the equations can be solved for  $B_2(p)$ . Taking the inverse transform we obtain

$$b_2(t) = -\frac{1}{2}\gamma(t-l/c)e^{-(t-l/c)\gamma/2} U(t-l/c). \quad (\text{A15})$$

Therefore,  $|\psi_2\rangle$  is not excited until the emission from one atom reaches the other atom. The energy that remains in the atomic excitation is  $(|b_1|^2 + |b_2|^2)\hbar\omega$ . For a general value of  $l$ , the decay is not exponential, and therefore the decay rate is not uniquely defined. However, we can define an effective decay rate  $\gamma'$  as the inverse of the time at which  $|b_1|^2 + |b_2|^2 = 1/e$ . This function  $\gamma'$  is plotted in Fig. 3. It is seen that the decay is superradiant for  $l < c/2\gamma$ , and that it is the normal decay for  $l > c/\gamma$ .

<sup>1</sup>R. H. Dicke, Phys. Rev. **93**, 99 (1954); *Proceedings of the Third Quantum Electronics Conference, Paris*, 1963, edited by N. Grivet and P. Bloembergen (Columbia U. P., New York, 1964), p. 35.

<sup>2</sup>F. Bloch, Phys. Rev. **70**, 460 (1946).

<sup>3</sup>R. P. Feynman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. **28**, 49 (1957).

<sup>4</sup>A. Abragam, *The Principles of Nuclear Magnetism* (Oxford U. P., London, 1961).

<sup>5</sup>F. T. Arecchi, E. Courtens, R. Gilmore, and H. Thomas (unpublished).

<sup>6</sup>This assumes a large Fresnel number,  $A/4\lambda l \gg 1$ , where  $l$  is the rod length. This is the usual case in optical experiments. For a detailed derivation see, for instance, I. D. Abella, N. A. Kurmit, and S. R. Hartmann, Phys. Rev. **141**, 391 (1966), Appendix C.

<sup>7</sup>See, for instance, D. L. Dexter and R. S. Knox, *Excitons* (Interscience, New York, 1965), p. 124, where it is argued that for  $10^{24}$  atoms with a spontaneous lifetime of  $10^{-8}$  sec, superradiance predicts a decay time of  $10^{-32}$  sec, which corresponds to a linewidth of  $10^8$  GeV around the optical frequency of 1 eV.

<sup>8</sup>We use rationalized mks units.

<sup>9</sup>For simplicity, we consider a circularly polarized field acting on a circularly polarized transition.  $\mathcal{O}$  is the matrix element of the  $x$  component of the transition dipole moment. For an aligned system, radiating in one polarization only, and in the direction of maximum emission, the transition probability in a solid angle  $d\Omega$  around that direction is given by

$$\gamma \frac{d\Omega}{4\pi} = \frac{8\pi^2 \mathcal{O}^2}{\hbar\epsilon\lambda^3} \frac{d\Omega}{4\pi}$$

This gives an effective value of  $\gamma$  appropriate to use in Eq. (2) for such a system.

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<sup>14</sup>That  $\tau_c$  is the time after which the atoms cease to emit individually and start to emit collectively is also found from formal derivations, for instance: A. I. Alekseev, Yu. A. Vdovin, and V. M. Galitskii, Zh. Eksperim. i Teor. Fiz. **46**, 320 (1963) [Soviet Phys. JETP **19**, 220 (1964)].

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