

Quantum Statistical Theory of Superradiance. II*

R. Bonifacio and P. Schwendimann

*Laboratorio Consiglio Nazionale delle Ricerche di Fisica del Plasma ed Elettronica Quantistica,
Istituto di Fisica, Università di Milano, Milan, Italy*

and

Fritz Haake[†]

Lyman Laboratory, Harvard University, Cambridge, Massachusetts 02138

(Received 29 January 1971)

We discuss the solution of the “superradiance master equation” derived in a preceding paper. During the first few photon transient times the cooperative atomic decay goes through a non-adiabatic oscillatory regime. For later times the decay takes place monotonically in time with the electromagnetic field following it adiabatically. The emitted light pulse has different statistical properties for an incoherently and a coherently prepared “superradiant” atomic initial state. The former case is characterized by large quantum fluctuations and strong atom-atom and atom-field correlations. In the latter case quantum fluctuations are small and the system behaves essentially classically. By also solving for a class of coherently prepared intermediate initial states we show that large quantum fluctuations occur only if the initial total occupancy of the excited state differs from the total number of atoms at most by a number of order unity.

I. INTRODUCTION

In a previous paper,¹ hereafter referred to as I, we derived a simple equation of motion for the density operator of an ensemble of many two-level atoms describing the cooperative decay of initial atomic excitation. This “superradiance master equation” has been shown to be valid under the following conditions: (i) The “active” volume filled by the atoms has a pencil shape, i. e., is very thin compared to its length. (ii) All atoms are identical. (iii) Any incoherent decay process—caused, for example, by nonradiative relaxation—is so slow that the atoms do not get out of phase with each other before the cooperative decay—caused by the coherent interaction with the electromagnetic field—is over. (iv) The length of the active volume is smaller than a self-consistently determined critical value, in order to guarantee that the envelope of the emitted light pulse is essentially constant over the length of the sample; intuitively speaking, this means that the atoms radiate as a single macroscopic dipole rather than seeing and producing different values of the field at different points of the sample. (v) At $t=0$, the field is in the vacuum state and the ensemble of atoms in some excited state with energy $E/\hbar\omega \gg 1$. To our knowledge, conditions (iii)–(v) are common to all of the numerous recent treatments of superradiance.² They characterize a physical situation in which one can expect spontaneous cooperative emission leading to the maximum possible peak value of the radiated intensity. We adopt condition (ii) for the sake of simplicity only.³ Condition (i) is a restriction not

made by other authors. It dispenses us from worrying about the angular distribution of the emitted radiation. As is well known, practically all of the energy initially stored in the atoms is radiated into the very small diffraction solid angle around the axial direction of the “pencil,” if condition (i) is met.⁴ We have shown in I that then the electromagnetic field in the active volume can be treated in terms of a single mode (end-fire mode) with a wave vector in the direction of the pencil and a frequency equal to the transition frequency of the two-level atoms. This enormous simplification of the problem is not possible for any other shape of the active volume. We are generously rewarded for restricting ourselves to this simple case by the fact that the resulting superradiance master equation is simple enough to be exactly solvable. In particular, its solution allows evaluation of the statistic properties of the emitted pulse, and thus control of the accuracy of semiclassical approximations on which most treatments of superradiance have been based up to now.

In this paper we present and discuss the solution of the superradiance master equation for various atomic initial states. The most important results are the following:

(a) For a “fully excited” atomic initial state (all atoms in the upper state, as realized, for example, by first preparing the atoms in the ground state and then shining the system with a classical “ π pulse”; more generally, for an ensemble of atoms in equilibrium with a completely incoherent pump mechanism) the emitted intensity $I(t)$ is at all times appreciably smaller than what a semiclassical rate

equation^{1,4} approach predicts. This discrepancy between quantal and semiclassical values is largest ($\sim 23\%$) near the peak intensity. The "normally ordered" intensity fluctuation, as measurable in a delayed photon coincidence experiment, is at all times much larger than it would be for a coherent state of the field. The dispersion of the total atomic energy $\langle R_3^2(t) \rangle - \langle R_3(t) \rangle^2$ which is neglected in the semiclassical rate equation approach assumes values of up to order N^2 near the maximum of the pulse. There are, at all times, large atom-atom and atom-field correlations. All of these results are found to be independent of or at least very insensitive to the number of atoms N , once N is large.

(b) For a "superradiant" atomic initial state (either the Dicke $|\frac{1}{2}N, 0\rangle$ state or, more generally and realistically, for a state prepared from the ground state by a classical $\frac{1}{2}\pi$ pulse) excellent agreement with the semiclassical description is found. In particular, we have $I(t) = I_{\text{class}}(t)$ to within at least 0.5%. The fluctuations of the emitted intensity and of the atomic energy are small. During most of the high-intensity region of the pulse the field may be considered to be in a coherent state, to within an accuracy $\sim 1/N$. Of course, these results can be understood intuitively to be due to the presence of a macroscopic initial value of a "transverse atomic dipole moment."

(c) If the atoms are prepared from the ground state by a classical " $2|\gamma|$ pulse" with $\frac{1}{2}\pi \leq 2|\gamma| \leq \pi$ (the angle $2|\gamma|$ may be looked upon as the classical Bloch angle; in our context it parametrizes a class of atomic states; see Sec. III), the behavior of the system ranges continuously between the two limits discussed above. However, the "classical" behavior prevails for all values of $2|\gamma|$ except those very close to the "fully excited" limit $2|\gamma| = \pi$. The "transition" value of $|\gamma|$ corresponds to a total occupancy of the upper state differing from N by a number of order unity.

(d) As we had already inferred from general arguments and now read off the explicit solution of the superradiance master equation, the emitted field follows the decay of the atoms adiabatically for times greater than the photon transient time, $t \gg \kappa^{-1}$. However for $t \lesssim \kappa^{-1}$ we find a distinctly non-adiabatic behavior of the system. Unfortunately, the non-Markovian effects taking place during this nonadiabatic regime are difficult to observe experimentally, as they have to be disentangled from the preparation process whose duration is just at least a photon transient time, too.

For the sake of the reader's convenience let us write down those results of I which will be needed in the following. We use the same notations as in I: $\kappa^{-1} = 2l/c$ equals the photon transient time; I_1 is the emission probability per unit time and unit

solid angle of an isolated atom times the diffraction solid angle of the end-fire mode; $R^\pm = \sum_{i=1}^N R_i^\pm e^{\pm ikr_i}$ are the total atomic dipole operators composed of raising (R_i^+) and lowering (R_i^-) operators for the individual atoms; k is the wave vector of the end-fire mode; $R_3 = \sum_{i=1}^N R_{3i}$ equals the total atomic energy operator apart from factor $\hbar\omega$; the R^α obey angular momentum commutation relations: $[R_3, R^\pm] = \pm R^\pm$; $[R^+, R^-] = 2R_3$; l is the length of the active volume; $l_{\text{thr}} = 4\pi(\gamma_1/\gamma)/\lambda^2\rho$ is the lower bound for l ; λ is the wavelength of the end-fire mode; γ is the natural linewidth of an isolated atom; γ_1 equals γ + collision or lattice broadening; ρ is the number density of atoms in the sample; $l_c = (4\pi c/\gamma\lambda^2\rho)^{1/2}$ equals the upper bound for l ; $g = (c\lambda^2\gamma/8\pi V)^{1/2}$ equals the atom-field coupling constant with V the volume of sample; a, a^\dagger are the annihilation and creation operators for photons in the end-fire mode.

Then conditions (iii) and (iv) above read

$$l_{\text{thr}} \ll l \ll l_c, \quad (1.1)$$

and the atomic density operator $W_A(t)$ obeys the superradiance master equation

$$\begin{aligned} \dot{W}_A(t) = \int_0^t ds \frac{1}{2} I_1 \kappa e^{-\kappa s} \{ [R^-, W_A(t-s) R^+] \\ + [R^- W_A(t-s), R^+] \}. \end{aligned} \quad (1.2)$$

Once Eq. (1.2) is solved, expectation values of field operators can be evaluated as

$$\begin{aligned} \langle a^\dagger a^m(t) \rangle = \left(-i \frac{g}{\kappa} \right)^{l+m} (-1)^l (l+m) \\ \times \int_0^t ds \kappa e^{-(l+m)\kappa s} (e^{\kappa s} - 1)^{l+m-1} \langle R^{+l} R^{-m}(t-s) \rangle. \end{aligned} \quad (1.3)$$

For large times, $t \gg \kappa^{-1}$, the theory constituted by (1.2) and (1.3) can be replaced by its Markovian version:

$$\dot{W}_A(t) = \frac{1}{2} I_1 \{ [R^-, W_A(t) R^+] + [R^- W_A(t), R^+] \} \quad (1.2')$$

and

$$\langle a^\dagger a^m(t) \rangle = (-i g/\kappa)^{l+m} (-1)^l \langle R^{+l} R^{-m}(t) \rangle. \quad (1.3')$$

If Eq. (1.2') is solved by suppressing fluctuations of the total atomic energy

$$\langle R_3^2(t) \rangle - \langle R_3(t) \rangle^2 = 0, \quad (1.4)$$

then the results of the semiclassical rate equation approach are recovered. Then the emitted intensity $I(t) = 2\kappa \langle a^\dagger a(t) \rangle$ has the well-known hyperbolic secant form

$$I(t) = I_1 \left[\frac{1}{2}(N+1) \right]^2 \text{sech}^2 \left[(t - t_m) / \tau_p \right], \quad (1.5)$$

with the pulse width

$$\tau_p^{-1} = \frac{1}{2} I_1 (N+1) \quad (1.6)$$

and a time t_m of maximal intensity which depends on the initial condition.

II. SOLUTION OF SUPERRADIANCE MASTER EQUATION

As we are finally interested in calculating normally ordered field expectation values $\langle a^{r_l} a^m(t) \rangle$ we have to solve the superradiance master equation only to the extent needed for the evaluation of the "corresponding" quantities $\langle R^{+l} R^{-m}(t) \rangle$. Therefore, and in view of the obvious fact that Eqs. (1.2) and (1.2') conserve total angular momentum it is convenient to use the angular momentum representation for the atomic density operator⁵:

$$W_A(t) = \sum_{\alpha, \alpha'} \sum_{r, r'=0}^{N/2} \sum_{m=-r}^{+r} \sum_{m'=-r'}^{+r'} W_{\alpha r m; \alpha' r' m'}(t) \times |\alpha r m\rangle \langle \alpha' r' m'|, \quad (2.1)$$

with

$$R_3 |\alpha r m\rangle = m |\alpha r m\rangle, \quad (2.2)$$

$$R^{\pm} |\alpha r m\rangle = [(r \mp m)(r \pm m + 1)]^{1/2} |\alpha r m \pm 1\rangle.$$

The "diagonal" expectation values $\langle R^{+l} R^{-l}(t) \rangle$ and $\langle R_3^l(t) \rangle$ —the former of which are of special importance because they determine the outcome of photon counting experiments—can be expressed in terms of the diagonal matrix elements $W_{\alpha r m; \alpha r m}(t)$ as

$$\langle R^{+l} R^{-l}(t) \rangle = \sum_{r, m} g(r, m) g(r, m-1) \cdots g(r, m-l+1) \times \sum_{\alpha} W_{\alpha r m; \alpha r m}(t) \\ \equiv \sum_{r, m} g(r, m) g(r, m-1) \cdots g(r, m-l+1) \times p(r, m, t), \quad (2.3)$$

$$\langle R_3^l(t) \rangle = \sum_{r, m} m^l p(r, m, t), \quad (2.4)$$

with $g(r, m) = (r+m)(r-m+1)$.

Conservation of angular momentum by the master equation (1.2) now express itself in the fact that the resulting equations of motion for the diagonal matrix elements $p(r, m, t)$ fall in separate blocks for each value of the "cooperation" quantum number.⁶ We get from Eq. (1.2)

$$\dot{p}(r, m, t) = I_1 \int_0^t ds \kappa e^{-\kappa s} [g(r, m+1) p(r, m+1, t-s) - g(r, m) p(r, m, t-s)], \quad (2.5)$$

and from Eq. (1.2')

$$\dot{p}(r, m, t) = I_1 [g(r, m+1) p(r, m+1, t) - g(r, m) p(r, m, t)]. \quad (2.5')$$

These equations describe one-step downward transitions between the collective energy levels m . Upward transitions are forbidden in the superradiance limit (1.1) (no reaction of the photons on the atoms is allowed). There are several standard methods available to solve the "recurrence relations" (2.5). The technically simplest one turns out to be a numerical procedure. We shall present the results thus obtained in Secs. IV–VI. However, an analytical solution may be easily obtained, too, and is best suited for a general discussion.

To construct the analytical solution of (2.5) and (2.5') we first observe that the linearity of these equations allows the introduction of a time evolution matrix according to

$$p(r, m, t) = \sum_{n=m}^r V_{nm}(r, t) p(r, n, 0), \quad (2.6)$$

$$V_{nm}(r, 0) = \delta_{nm}.$$

The Laplace transform of $V_{nm}(r, t)$,

$$V_{nm}(r, z) = \int_0^t dt e^{-zt} V_{nm}(r, t), \quad (2.7)$$

may be found from the Laplace-transformed recurrence relations (2.5) by iteration to read

$$V_{nm}(r, z) = \frac{\kappa + z}{\kappa I_1 g(r, m)} \prod_{l=m}^n \frac{\kappa I_1 g(r, l)}{z(\kappa + z) + \kappa I_1 g(r, l)} \quad (2.8)$$

or, from (2.5'),

$$V_{nm}(r, z) = \frac{1}{I_1 g(r, m)} \prod_{l=m}^n \frac{I_1 g(r, l)}{z + I_1 g(r, l)}. \quad (2.8')$$

The absence of terms with $n < m$ or, equivalently, the vanishing of the time evolution matrix for $n < m$ shows again that probability flows only down the collective energy ladder. The behavior of the time-dependent $V_{nm}(r, t)$ is characterized by the singularities of $V_{nm}(r, z)$. The latter are poles at

$$Z_l^{\pm} = -\frac{1}{2} \kappa \{1 \pm [1 - 4I_1 g(r, l)/\kappa]^{1/2}\} \quad (2.9)$$

and

$$Z_l = -I_1 g(r, l). \quad (2.9')$$

Note that because of $g(r, m) \geq 0$ we have $\text{Re} Z_l^{\pm} \leq 0$. The poles therefore give rise to decaying exponentials $\exp(Z_l^{\pm} t)$ and $\exp(Z_l t)$, respectively, in $V_{nm}(r, t)$. According to the symmetry property $g(r, l) = g(r, -l+1)$ some of the poles may coincide in pairs whereupon the corresponding exponentials acquire first-order polynomials in t as factors. The termination of the collective energy ladder at $m = -r$ shows up in $g(r, -r) = 0$ which entails $Z_{-r} = Z_{-r} = 0$. This leads to an accumulation of probability in the ground level as time elapses. Indeed, this pole is the only one to contribute to the station-

ary solution

$$V_{nm}(t \rightarrow +\infty) = \lim_{z \rightarrow 0^+} z V_{nm}(r, z) = \delta_{m, -r}. \quad (2.10)$$

Together with probability conservation

$$\sum_{m=-r}^{+r} p(r, m, t) = 1$$

and the definition (2.7), this gives

$$p(r, m, t \rightarrow \infty) = \delta_{m, -r} \quad (2.11)$$

independent of the initial probabilities.

Finally, we are now able to compare explicitly the results of the non-Markovian and the Markovian treatment. We had already concluded from general arguments that both theories give identical results for times larger than the photon transient time κ^{-1} . To see that now, explicitly observe that for $t \gg \kappa^{-1}$ only those exponentials in $V_{nm}(r, t)$ or $p(r, m, t)$ survive whose damping constants are small as compared to κ . The latter are indeed $-Z_i^{\pm} = -Z_i = g(r, l)I_1 \ll \kappa$. However, for times $t \leq \kappa^{-1}$ the respective predictions differ drastically. In this non-adiabatic regime none of the exponentials $\exp(Z_i^{\pm} t)$ is negligible. In particular, for $\kappa < 4I_1 g(r, l)$ or $\kappa < 2g[2g(r, l)]^{1/2}$, the non-Markovian poles Z_i^{\pm} develop imaginary parts. This implies, as a feature quite typical for non-Markovian effects, that some observables of the system need not decay monotonically in time but may rather display a damped oscillation behavior. Note that the above condition for this to happen is compatible with the superradiance limit (1.1) for $r \lesssim \frac{1}{2}N$, $l \ll r$. Summarizing this discussion we may conclude that the electromagnetic field, starting from the initial vacuum state, catches up with the motion of the atoms within a time of order κ^{-1} and then follows the latter adiabatically.⁷

III. PREPARATION OF ATOMIC INITIAL STATE

Let us briefly discuss two excitation mechanisms which can be used to prepare our system to different initial states. These mechanisms have to be designed to pump energy into the atoms without producing photons in the end-fire mode and, very importantly, without interfering with the cooperative decay process. The latter requirement means that the characteristic times of the pump must be much smaller than the duration of the cooperative decay.

First, consider an arbitrary but incoherent pump leaving the atoms in an uncorrelated equilibrium state

$$W_A(0) = \prod_{i=1}^N \frac{\exp[(\hbar\omega/KT_{\text{eff}})R_{3i}]}{2\cosh(\hbar\omega/2KT_{\text{eff}})}. \quad (3.1)$$

The effective temperature T_{eff} characterizing this equilibrium with the pump may be negative to ac-

count for an eventually positive inversion for the N two-level atoms. As has already been pointed out by Dicke,⁸ the inversion R_3 (or the atomic energy $\hbar\omega R_3$) has an extremely small relative dispersion in the state (3.1) for $N \gg 1$:

$$\langle R_3^2 \rangle - \langle R_3 \rangle^2 = \frac{1}{4}N \operatorname{sech}^2 \frac{\hbar\omega}{2KT_{\text{eff}}} \leq \frac{1}{4}N. \quad (3.2)$$

Therefore, up to corrections of order $1/\sqrt{N}$ we can replace the canonical ensemble (3.1) by a micro-canonical ensemble

$$W_A(0) = Z^{-1} \sum_{\alpha} \sum_{r=|m|}^{N/2} |\alpha r m\rangle \langle \alpha r m|, \quad (3.3)$$

with energy $\hbar\omega m$ according to

$$m = \langle R_3 \rangle = -\frac{1}{2}N \tanh \frac{\hbar\omega}{2KT_{\text{eff}}} \geq 0 \text{ for } T_{\text{eff}} \leq 0. \quad (3.4)$$

Moreover, Dicke⁸ has shown that for energies of interest in the present context, $m^2 \gg \frac{1}{2}N \gg 1$, Eq. (3.3) may be replaced with

$$W_A(0) = |r, m\rangle \langle r, m|, \quad r = |m|, \quad m \text{ as above} \quad (3.5)$$

because the state with the smallest possible cooperation quantum number $r = |m|$ has an overwhelming statistical weight in (3.3). Hence an incoherently prepared atomic initial state may be specified in terms of the probabilities $p(r, m)$ introduced in Sec. II as

$$p(r', m') = \delta_{r, |m|} \delta_{m', m} \quad (3.6)$$

with m as in (3.4). In particular, for small negative temperatures, $|\hbar\omega/KT_{\text{eff}}| \rightarrow 0$, the atoms can be considered to be in the fully excited state $r = m = \frac{1}{2}N$.

As a completely different situation let us now encounter a coherent pump. We imagine all atoms to be in the ground state before a pulse of resonant coherent light runs along the pencil-shaped cavity. The pulse is assumed so strong that it can be treated as a constant classical source acting on the atoms for a time T (which, again, has to be short compared to the characteristic times of the superradiance process). The Hamiltonian for such a simple process reads

$$H = \hbar g(\alpha R^+ + \alpha R^-). \quad (3.7)$$

The complex quantity α defines the fixed amplitude and phase of the pump field. The atomic state generated by a classical field according to the Hamiltonian (3.7) is the formal analog of the coherent state of the electromagnetic field generated from the vacuum by a prescribed classical current. The evaluation of this "quasiclassical" state of the system is a straightforward algebraic affair⁹:

$$|\psi(T)\rangle = e^{-iHT/\hbar} \prod_{i=1}^N |-\rangle_i \quad (3.8)$$

$$= \prod_{i=1}^N (\cos|\gamma| |-\rangle_i + \sin|\gamma| e^{i\varphi} |+\rangle_i), \quad (3.9)$$

with

$$\gamma = -igT\alpha = |\gamma| e^{i\varphi}.$$

The meaning of the angles $|\gamma|$ and φ is elucidated from the expectation values $\langle x \rangle = \langle \psi(T) | x | \psi(T) \rangle$:

$$\begin{aligned} \langle R^x \rangle &= \frac{1}{2} N \sin^2 |\gamma| e^{i\varphi}, \\ \langle R_z \rangle &= -\frac{1}{2} N \cos 2|\gamma|. \end{aligned} \quad (3.10)$$

Hence $2|\gamma|$ is the angle between the classical vector $\langle R \rangle$ and its third component, whereas φ fixes the phase of the transverse component. Note that $|\psi(T)\rangle$, unlike an $|r, m\rangle$ state, is an uncorrelated state in the sense that it factors into single-atom wave functions.

Observe now that the quasiclassical state $|\psi(T)\rangle$ is completely symmetric in all atoms. We therefore suspect and indeed easily verify that it may be represented as a linear combination of the energy eigenstates $|r, m\rangle$ with $r = \frac{1}{2}N$ which have the same symmetry:

$$\begin{aligned} |\psi(T)\rangle &= \sum_{m=-N/2}^{+N/2} \binom{N}{\frac{1}{2}N-m}^{1/2} (\cos|\gamma|)^{N/2-m} \\ &\quad \times (\sin|\gamma| e^{i\varphi})^{N/2+m} |\frac{1}{2}N, m\rangle. \end{aligned} \quad (3.11)$$

Taking the state $|\psi(T)\rangle$ as initial state of the atoms we have

$$\begin{aligned} W_A(0) &= |\psi(T)\rangle \langle \psi(T)| \\ &= \sum_{m=-N/2}^{+N/2} p(\frac{1}{2}N, m, 0) |\frac{1}{2}N, m\rangle \langle \frac{1}{2}N, m| \\ &\quad + \sum_{m \neq m'} W_{N/2, m; N/2, m'}(0) |\frac{1}{2}N, m\rangle \langle \frac{1}{2}N, m'|, \end{aligned} \quad (3.12)$$

with the probabilities

$$p(\frac{1}{2}N, m, 0) = \binom{N}{\frac{1}{2}N-m} (\cos^2|\gamma|)^{N/2-m} (\sin^2|\gamma|)^{N/2+m}, \quad (3.13)$$

and the nondiagonal elements ($m \neq m'$)

$$\begin{aligned} W_{N/2, m; N/2, m'}(0) &= \left[\binom{N}{\frac{1}{2}N-m} \binom{N}{\frac{1}{2}N-m'} \right]^{1/2} \\ &\quad \times (\cos|\gamma|)^{N-m-m'} (\sin|\gamma|)^{N+m+m'} e^{i\varphi(m-m')}. \end{aligned} \quad (3.14)$$

Let us note that if the pump pulse has a fixed amplitude $\propto |\gamma|$ but a random phase φ , the nondiagonal elements (3.14) will vanish. The probabilities $p(\frac{1}{2}N, m)$, however, are phase independent. Therefore, an excitation by an "unphased" and a "phased" pulse with equal amplitude and duration will produce the same behavior of the system as far as "diagonal" quantities are concerned.

It is well known and easy to see from (3.13) that

an excitation by what is usually called a π pulse which, in our notation, corresponds to $|\gamma| = \frac{1}{2}\pi$ leaves the atoms in the fully excited state $p(r, m, 0) = \delta_{r, N/2} \delta_{m, N/2}$. On the other hand, a " $\frac{1}{2}\pi$ pulse" (i. e., $|\gamma| = \frac{1}{4}\pi$) produces a binomial probability distribution

$$p(r, m, 0) = \delta_{r, N/2} \binom{N}{\frac{1}{2}N-m},$$

with mean value $\langle m \rangle = \langle R_z \rangle = 0$. For large N the binomial distribution is sharply peaked around the mean value so that it may eventually be replaced with $p(r, m) = \delta_{r, N/2} \delta_{m, 0}$ which corresponds to Dicke's superradiant state. In this sense the Dicke state $|r, m=0\rangle$ can be said to be realizable. However, as the binomial distribution is not more difficult to cope with than the "Dicke" one, we base our numerical work on the former.

Let us point out that the nonadiabatic regime of the cooperative decay process will be unobservable if the initial state is prepared by a classical light pulse. The minimal duration of the excitation is given by the travel time of the pulse across the length of the sample which is just the characteristic time of the nonadiabatic process (see Sec. II).

IV. RESULTS FOR FULLY EXCITED INITIAL STATE

Restricting ourselves to the adiabatic regime we

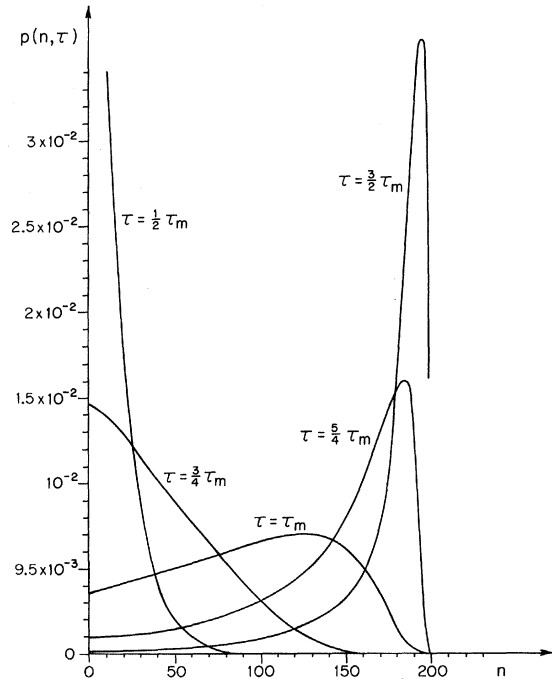


FIG. 1. Probability distribution $p(n, \tau)$ at various times $\tau = t/\tau_p$ (τ_p : pulse width; $\tau_m = t_m/\tau_p$: time of pulse maximum) for values of $n = r - m$ ranging from 0 to 200, starting from initial condition $p(n, 0) = \delta_{n, 0}$.

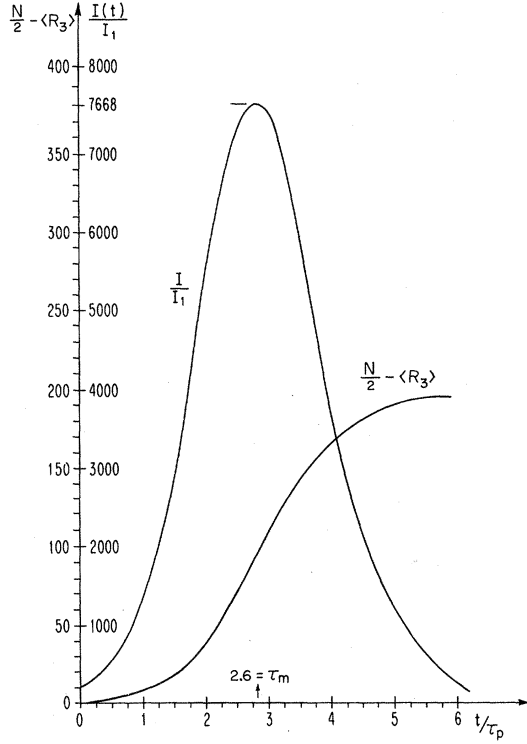


FIG. 2. Radiated intensity I/I_1 and numbers of decayed atoms $N_0 = r - \langle R_3 \rangle$ for $N = 200$ initially excited atoms.

have numerically integrated Eq. (2.5') for the initial condition $p(r, m, 0) = \delta_{r,m}$ with $r = 100$. According to the considerations of Sec. III this initial probability distribution corresponds to either an incoherently prepared system of N atoms with cooperation quantum number $r = m = 100 \leq \frac{1}{2}N$ or to a coherently prepared fully excited system with $N = 2r = 200$. In the following we shall refer to this initial condition as "fully excited initial state." The resulting probability distribution $p(r, m, t)$ is plotted in Fig. 1 as a function of $n = r - m$ at different times. The quantity $n = r - m$ counts the number of atoms which have decayed to the fundamental level and is therefore related to the number of emitted photons. We see that $P_n(t)$ first broadens to resemble a Bose-Einstein distribution and finally be-

comes again a sharply peaked function tending asymptotically to $p_n(t \rightarrow \infty) = \delta_{n,2r}$ when all atoms have decayed to the ground level.

In Fig. 2 (see also Table I) we display the resulting behavior of the radiated intensity $I(t) = 2\kappa \langle a^\dagger a(t) \rangle = I_1 \langle R^+ R^-(t) \rangle$ and of the number of atoms in the ground state $N_0(t) = r - \langle R_3(t) \rangle$. Observe that the shape of $I(t)$ is similar to that of the classical hyperbolic secant. In particular, the time t_m after which the intensity reaches its maximum value is found to be $t_m = 2.8\tau_p$, i. e., within some 5% of the classical prediction $\tau_m = 2.65\tau_p$. The most interesting difference from the classical behavior consists in the fact that the maximum value of the intensity is only about 76.7% of the corresponding classical value r^2 . This feature of the quantum-mechanical theory was confirmed by repeating the calculation for $N = 10^4$ atoms (see Table II). It is easy to see that the quantum mechanically calculated intensity is always smaller than the classically calculated one¹⁰:

$$\begin{aligned} I(t) &= I_1 \langle R^+ R^-(t) \rangle = \sum_{m=-r}^{+r} g(r, m) p(r, m, t) \\ &= I_1 \sum_{m=-r}^{+r} [r(r+1) - m^2 + m] p(r, m, t) \\ &= I_{\text{class}}(t) - [\langle R_3^2(t) \rangle - \langle R_3(t) \rangle^2]. \end{aligned} \quad (4.1)$$

Therefore $I(t) < I_{\text{class}}(t)$, apart from the initial and final situation where the system is in an energy eigenstate. The 23.3% difference observed in the superradiant domain of the pulse for both $N = 2 \times 10^2$, 10^4 indicates that the peak value of the dispersion of R_3 is of order r^2 and surprisingly insensitive to the value of r . A plot of the latter, namely,

$$\sigma^2(R_3) = [\langle R_3^2(t) \rangle - \langle R_3(t) \rangle^2] / r^2 \quad (4.2)$$

as a function of time is shown in Fig. 3. Amazingly enough, the classical approximation is seen to be worst near the maximum of the emitted pulse. We shall show in Sec. VII that the large dispersion of R_3 is due to large atom-atom energy correlations.

The normally ordered intensity fluctuation

$$\sigma_n^2(I) \equiv \frac{\langle a^\dagger a^\dagger a a(t) \rangle - \langle a^\dagger a(t) \rangle^2}{\langle a^\dagger a(t) \rangle^2}$$

TABLE I. Numerical values of various quantities for incoherent pumping when the initial state is an $|r, m\rangle$ state with $r = m = \frac{1}{2}N = 10^2$. τ is defined as t/τ_p .

τ	0.5×10^{-2}	0.25	0.50	1.00	2.00	$\tau_m = 2.80$	3.00	4.00	6.40
$\langle I \rangle / I_1$	200	327	532	1340	5406	7758	7580	3965	132
$\sigma_n^2(I)$	0.99	0.98	0.96	0.84	0.33	0.102	9.98×10^{-2}	0.5	
$N_0 = r - \langle R_3 \rangle$	0	0.64	1.7	6	37	92.6	108	167	198
$\sigma^2(R_3)$	10^{-6}	10^{-4}	5×10^{-4}	4.1×10^{-3}	8.4×10^{-2}	0.229	0.24	0.14	2×10^{-2}

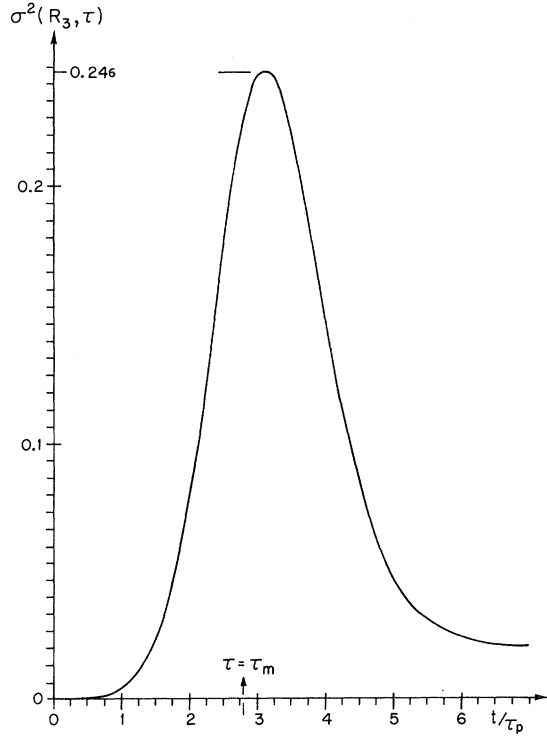


FIG. 3. Dispersion of total inversion (atomic energy) $\sigma^2(R_3) = (\langle R_3^2 \rangle - \langle R_3 \rangle^2) / r^2$ for $N = 2r = 200$ initially excited atoms.

$$= \frac{\langle R^* R^* R^- R^-(t) \rangle - \langle R^* R^-(t) \rangle^2}{\langle R^* R^-(t) \rangle^2}, \quad (4.3)$$

which can be measured in a photon-count experiment is plotted as a function of time in Fig. 4. It is seen to go through a minimum in the high-in-

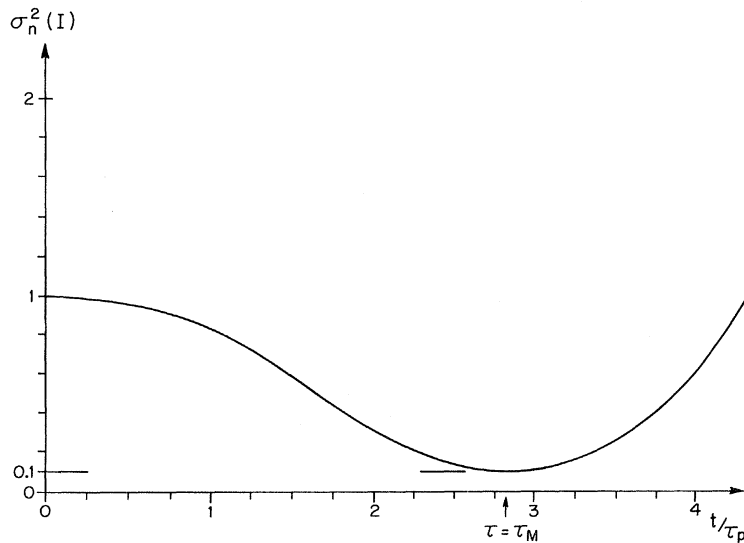


FIG. 4. Normally ordered intensity fluctuation $\sigma_n^2(I)$ for $N = 200$ initially excited atoms.

TABLE II. Same as Table I with $r = m = 5 \times 10^3$ for $\tau = \tau_m = \ln(2r)^{1/2}$. Note the coincidence with the previous case for the relative dispersions.

	$\tau_m = 4.78$
$\langle I \rangle$	2×10^7
$\sigma_n^2(I)$	0.102
$N_0 = r - \langle R_3 \rangle$	4757
$\sigma^2(R_3)$	0.227

tensity region. The minimum value 0.1 of $\sigma_n^2(I)$ is not in agreement with the values obtained for Poisson statistics [$\sigma_n^2(I) = 0$] or Bose statistics [$\sigma_n^2(I) = 1$]. It is found to be insensitive to the number of atoms by repeating the calculation for $r = 10^4$ (see Table II).

The above results, notably those concerning the insensitivity of the decay and emission process to changes of r (once r is large), allow the conclusion that the cooperative decay of an initially "fully excited" atomic state cannot satisfactorily be described in classical terms.

V. RESULTS FOR SUPERRADIANT INITIAL STATE

As follows from the considerations in Sec. III, a coherent excitation pulse with $2|\gamma| = \frac{1}{2}\pi$ prepares the atoms in a coherent superposition of the excited and ground levels with equal probabilities. The resulting binomial probability distribution $p(r, m, 0) \equiv p(n)$, $n = \frac{1}{2}N - m$,

$$p(n) = 2^{-N} \binom{N}{n} \quad (5.1)$$

is sharply peaked around the mean value $\langle n \rangle = \frac{1}{2}N$ ($\langle m \rangle = 0$) with a relative dispersion $1/\sqrt{N}$, and there-

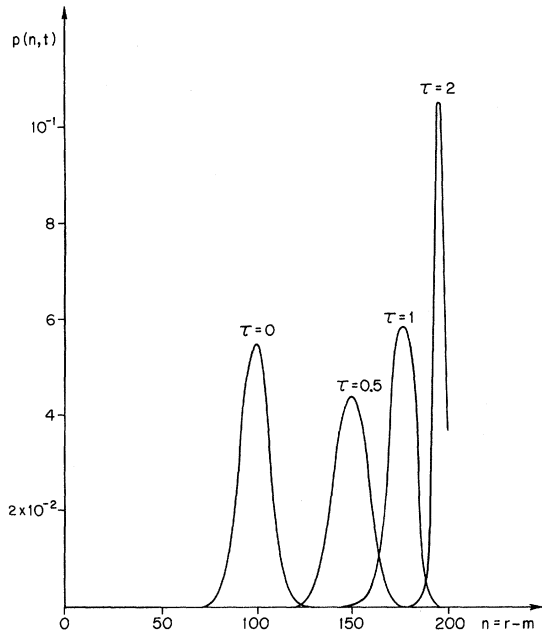


FIG. 5. Same as Fig. 1 for $N=200$ coherently excited atoms ($2|\gamma| = \frac{1}{2}\pi$).

fore is practically identical to Dicke's superradiant state $p(r, m, 0) = \delta_{r, N/2} \delta_{m, 0}$.

In Fig. 5, we show the time-dependent $p(n, t)$ as obtained by solving Eq. (2.5') numerically for the initial condition (5.1) and $N=200$ atoms. It is interesting to note that for $t \geq 0$, $p(n, t)$ does not remain a binomial distribution. This is a consequence of the obvious fact that the master equation (2.3) does not preserve the "quasiclassical" state $|\psi(T)\rangle$. The dispersion of n calculated with the solution $p(n, t)$ is always larger than the dispersion of a binomial distribution with equal mean value $\langle n \rangle$.

The intensity $I(t)$ for the present $p(n, t)$ is plotted in Fig. 6. In contrast to what we observed for the fully excited initial state in Sec. IV the agreement with the semiclassical hyperbolic secant behavior is excellent. By inspection of the expression (4.1) for $I(t) - I_{\text{class}}(t)$ and the numerical values for $\sigma^2(R_3)$ given in Table III it may be seen that $I(t)$ at no time differs from $I_{\text{class}}(t)$ by more than 0.5%.¹¹ Also, the value $\frac{1}{2}I(0)$ is reached after $t/\tau_p = 0.877$, in agreement with the classical prediction $t/\tau_p = \ln(1 + \sqrt{2})$ obtained from Eqs. (1.5) and (1.6) with $N=200$, $t_m = -\tau_p/N \approx 0$. The results for the normally ordered intensity fluctuation $\sigma_n^2(I)$, shown in Fig. 7 and Table III, also lead to the conclusion that the emitted field behaves classically. Except for large times ($t > 2\tau_p$), where $\langle a^\dagger a(t) \rangle^2$ tends to zero faster than $\langle a^\dagger a^\dagger a a(t) \rangle - \langle a^\dagger a(t) \rangle^2$, $|\sigma_n^2(I)|$ is found to be very small compared to unity. For $t \lesssim 0.5\tau_p$, i. e., during most of the high-intensity region of the

pulse, we even have $|\sigma_n^2(I)|^2$ of the order $1/N$, i. e., values certainly allowing to characterize the field as a "classical" one: To within an accuracy $1/N$ the field may be considered to be in a coherent state in that region.¹²

VI. RESULTS FOR INTERMEDIATE INITIAL STATES

In Secs. IV and V we have studied the behavior of the atom-field system for two initial states which may be looked upon as limiting cases of the more general state (3.11) and (3.13) with $\frac{1}{2}\pi \leq 2|\gamma| \leq \pi$. As the dynamical and statistical properties of the system are quite different for $2|\gamma| = \frac{1}{2}\pi$ and π , the question naturally arises how large the quantum fluctuations are for initial states with intermediate values of $2|\gamma|$. Therefore we have calculated the maximum value of $\sigma^2(R_3)$ and the minimum value of $\sigma_n^2(I)$ for various initial states with $2|\gamma|$ between $\frac{1}{2}\pi$ and π . Clearly, these quantities give a rough measure of how close to classical behavior the system gets during the pulse. The numerical results obtained with $N=2\gamma=10^3$ are listed in Fig. 8 and Table IV. We see that both $\sigma_n^2(I)|_{\text{min}}$ and $\sigma^2(R_3)|_{\text{max}}$ are very small compared to unity and, moreover, quite close to the corresponding values obtained for $2|\gamma| = \frac{1}{2}\pi$ over most of the range of $2|\gamma|$. Large quantum fluctuations occur only for $2|\gamma|$ very close to π . Using $\langle R_3(0) \rangle = -\frac{1}{2}N \cos 2|\gamma|$ we see that the transition takes place if the initial occupancy of the upper state $N_1(0) = \frac{1}{2}[N + 2\langle R_3(0) \rangle]$

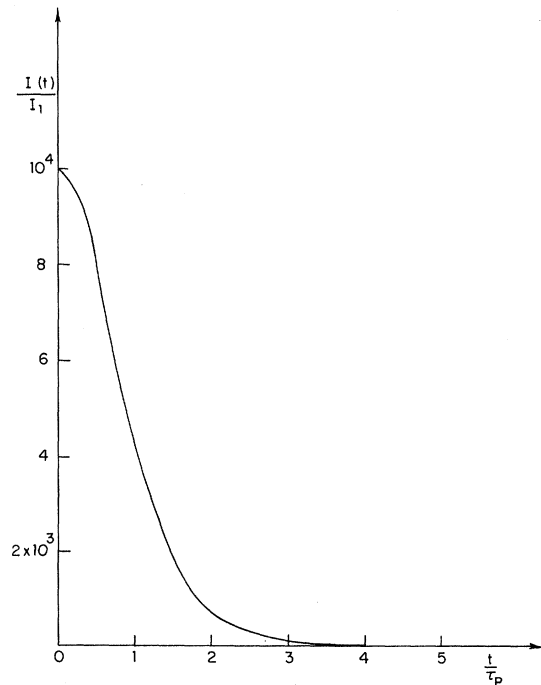


FIG. 6. Same as Fig. 2 for $N=200$ coherently excited atoms ($2|\gamma| = \frac{1}{2}\pi$).

TABLE III. Numerical results for coherent pumping when the initial state is a quasiclassical superradiant state ($\nu = 10^2$).

	0.5×10^{-2}	0.25	0.50	1.00	2.00	3.00	4.00	6.40
$\langle I \rangle$	10049	9441	7914	4271	735	104	14.3	0.12
$\sigma_n^2(t)$	-0.215×10^{-4}	-0.294×10^{-2}	0.7×10^{-3}	0.187×10^{-1}	0.745×10^{-1}	0.123	0.173	0.286
$N_0 = \nu - \langle R_3 \rangle$	99.5	123	145.3	175.5	196.2	199.4	199.9	199.999
$\sigma^2(R_3)$	0.51×10^{-2}	0.78×10^{-2}	0.81×10^{-2}	0.44×10^{-2}	0.6×10^{-3}	0.17×10^{-3}	0.13×10^{-3}	0.13×10^{-3}

differs from N by a number of order unity. For smaller values of $2|\gamma|$ or $N_1(0)$ the system displays only small quantum fluctuations and, as an important consequence, can be described correctly by the semiclassical approach [$\langle R_3^2(t) \rangle = \langle R_3(t) \rangle^2$]. Moreover, the completely classical approach using the Bloch equations is appropriate in this range, too. In order to see this we only have to recall that we have seen in I, Appendix B, that classical and semiclassical approaches lead to identical results for $N \gg 1$, and $N_1(0) \lesssim N - 1$.

VII. ATOM-ATOM AND ATOM-FIELD CORRELATIONS

We have seen in Sec. IV that the cooperative decay of N initially fully excited atoms is accompanied by large quantum fluctuations. In particular, the relative dispersion of the energy $\sigma^2(R_3) = I_{\text{class}}(t) - I(t)$, reaches a peak value of ~ 0.23 , which is rather insensitive to the number of atoms. It is easy to see that this is due to large atom-atom energy correlations. Indeed, recalling $R_3 = \sum_{i=1}^N R_{3i}$ we have, with $\Delta \equiv \sigma^2(R_3) \nu^2 = \langle R_3^2 \rangle - \langle R_3 \rangle^2$,

$$\Delta^2 = \sum_{i=1}^N \sigma_{ii} + \sum_{i \neq j} \sigma_{ij}, \tag{7.1}$$

with $\sigma_{ij} = \langle R_{3i} R_{3j} \rangle - \langle R_{3i} \rangle \langle R_{3j} \rangle$.

Now, because of the symmetry of both the superradiant master equations (2.2) and (2.3) and our initial conditions with respect to the exchange of two atoms, we have

$$\sigma_{ii} = \sigma, \quad \sigma_{ij} = \sigma_{ji} \equiv \sigma_{12} \quad \text{for } i \neq j,$$

and therefore

$$\Delta^2 = N\sigma + N(N-1)\sigma_{12}. \tag{7.2}$$

This shows that the total energy dispersion has contributions from single-spin fluctuations and spin-

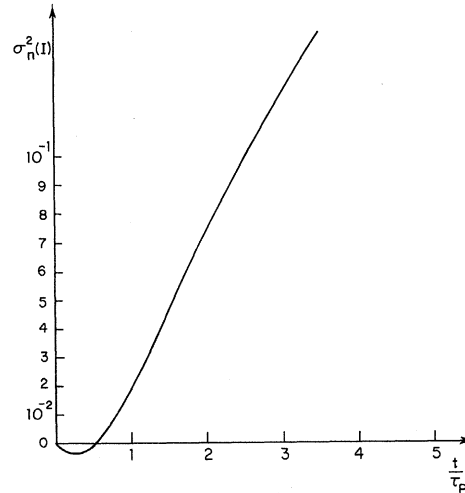


FIG. 7. Same as Fig. 4 for $N=200$ coherently excited atoms ($2|\gamma| = \frac{1}{2}\pi$).

TABLE IV. Numerical results for intermediate initial states prepared by $2|\gamma|$ pulse with $\frac{1}{2}\pi \leq 2|\gamma| \leq \pi$.

$2 \gamma $	$\sigma_{\max}^2(R_3)$	$\sigma_{n,\min}^2(I)$
$\frac{1}{2}\pi$	0.43×10^{-2}	0.25×10^{-3}
$\frac{5}{8}\pi$	0.58×10^{-2}	0.26×10^{-3}
$\frac{3}{4}\pi$	0.94×10^{-2}	0.20×10^{-3}
$\frac{7}{8}\pi$	0.22×10^{-1}	0.92×10^{-3}
$\frac{15}{16}\pi$	0.6×10^{-1}	0.73×10^{-2}
$\frac{31}{32}\pi$	0.15	0.44×10^{-1}
π	0.25	0.1

spin energy correlations. Note that $\sigma \leq \frac{1}{4}$, $\sigma_{12} \leq \frac{1}{2}$ for the two-level atoms. Our observation that Δ^2 is of order N^2 therefore simply means that the correlation term σ_{12} is of order unity or, more precisely, near its maximum possible value $\frac{1}{2}$. In other words, spin-spin energy correlations completely dominate single-spin fluctuations in (7.2). We have already noted that the superradiance master equations (2.2) and (2.3) do not allow for an uncorrelated solution, i.e., for a $W_A(t)$ factorizing with respect to the individual atoms. We here see that, moreover, any attempt to solve the master equations approximately using a factorizing ansatz for $W_A(t)$ is bound to fall short of a proper description of the superradiant decay of initially fully excited atoms. Such an ansatz completely suppresses the correlation σ_{12} .

The importance of atom-atom correlations for the superradiant pulse also elucidates from a microscopic analysis of the radiated intensity

$$\frac{I(t)}{I_1} = \langle R^* R^- \rangle = \sum_i \langle R_i^* R_i^- \rangle + \sum_{i \neq j} \langle R_i^* R_j^- \rangle. \quad (7.3)$$

Observing that $\langle R_i^* R_i^- \rangle = N_{1i}$ is the population of the upper level of the i th atom and that, for symmetry

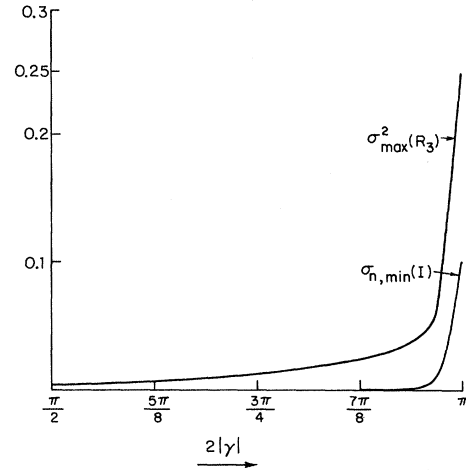


FIG. 8. Maximum value $\sigma_{\max}^2(R_3)$ of dispersion of total atomic energy and minimum value $\sigma_{n,\min}^2(I)$ of normally ordered intensity fluctuation for coherent excitation by $2|\gamma|$ pulses with varying value of $2|\gamma|$.

reasons, $\langle R_i^* R_j^- \rangle = r_{12} = r_{21} = r_{12}^*$, this may be written as

$$I(t)/I_1 = N_1(t) + N(N+1)r_{12}(t). \quad (7.4)$$

We see that $I(t)$ is made up by the ordinary spontaneous emission¹³ proportional to the total population $N_1(t)$ of the upper level and a term proportional to N^2 and the dipole-dipole correlation r_{12} . Evidently, as $N_1(t) \leq N$, the superradiant enhancement of the emitted intensity is due solely to the correlation term r_{12} . Let us note that r_{12} is space independent because it refers to the phase dipole operators $R_i^+(k) = R_i^+ e^{i k r_i}$. The correlation between the true dipole operators is space dependent. Consider the real quantity

$$r_{ij}(k) = \frac{1}{2} (\langle R_i^* R_j^- \rangle + \langle R_j^* R_i^- \rangle), \quad (7.5)$$

TABLE V. Coherence and correlation properties for different initial states. Field phase coherence \leftrightarrow atomic dipole incorrelation ($\langle R_i^* R_j^- \rangle = \langle R_i^* \rangle \langle R_j^- \rangle$); field amplitude coherence \leftrightarrow atomic energy incorrelation ($\langle R_{3i} R_{3j} \rangle = \langle R_{3i} \rangle \langle R_{3j} \rangle$).

Initial state	$\langle R^* \rangle \propto \langle a^\dagger \rangle$	$\langle R_{3i} R_{3j} \rangle - \langle R_{3i} \rangle \langle R_{3j} \rangle$	$\frac{\langle R_i^* R_j^- \rangle - \langle R_i^* \rangle \langle R_j^- \rangle}{N^2}$	$\langle a^\dagger R^- \rangle - \langle a^\dagger \rangle \langle R^- \rangle$	$\sigma_n^2(I)$
Fully excited	0	0.2 (at $t = t_m$)	$\langle R^* R^- \rangle / N^2$	$(g/\kappa) \langle R^* R^- \rangle$	0.1 (at $t = t_m$)
Coherent excitation by a $\frac{1}{2}\pi$ pulse with fixed phase and amplitude	$\sim \langle R^* R^- \rangle^{1/2}$	Very small	Very small	Very small	Very small
Coherent excitation by a $\frac{1}{2}\pi$ pulse with fixed amplitude and random phase.	0	Very small	$\langle R^* R^- \rangle / N^2$	$(g/\kappa) \langle R^* R^- \rangle$	Very small

where now R_i^\pm are the "true" nonphased operators. Expressing them in terms of their phased analogs and using (6.3) and (6.4), we get

$$r_{ij}(k) = \langle \langle R^+ R^- \rangle \rangle / N^2 \cos k(r_i - r_j) - O(1/N). \quad (7.6)$$

This shows the explicit dependence of the dipole correlation on the atomic positions when, by superradiant enhancement, $\langle R^+ R^- \rangle$ becomes of order N^2 .

It is also enlightening to look into the atom-field correlations accompanying the superradiant pulse. These can be discussed in terms of quantities as $\langle AF \rangle - \langle A \rangle \langle F \rangle$, where A, F are atomic and field operators, respectively. Consider, e.g.,¹⁴

$$c \equiv |\langle a^\dagger R^- \rangle - \langle a^\dagger \rangle \langle R^- \rangle| \\ = (g/\kappa) (\langle R^+ R^- \rangle - \langle R^+ \rangle \langle R^- \rangle). \quad (7.7)$$

It is easy to see that for the coherently prepared quasiclassical initial state (3.11)

$$c = (g/\kappa) \sin^4 |\gamma| \\ \text{and} \quad (7.8)$$

$$c_r \equiv c / \langle a^\dagger \rangle \langle R^- \rangle = 1/N \cos^2 |\gamma|.$$

This shows that the initial atom-field correlations are not significant for $|\gamma| = \frac{1}{4}\pi$ which indeed, as we have seen, gives rise to the "most classical" be-

havior of the system. On the other hand, for any incoherently prepared initial state we have $\langle R^+(t) \rangle = 0$, whereupon $c = (g/\kappa) \langle R^+ R^- \rangle = (1/2g) I(t)$. In this case, evidently, the radiated intensity provides a direct measure of the (extremely strong!) atom-field correlations.

The differences in the statistical behavior of our system caused by different initial conditions are summarized in Table V. This table may also be read as a confirmation of the general rule¹⁵ that phase and amplitude coherence exclude correlations between phased and unphased quantities, respectively and independently. We see, for instance, that phase incoherence ($\langle R^+ \rangle = 0$) implies correlation only between quantities involving phase as the dipole-dipole correlation but does not imply any correlation between the energies of different atoms. On the other hand the energy correlation for different atoms vanishes as long as the field has amplitude coherence [$\sigma_n^2(t) \approx 0$].

ACKNOWLEDGMENT

The authors express their gratitude to Dr. A. Airoidi, Milan, for the numerical solution of Eq. (2.5').

*Work supported in part by the Air Force Office of Scientific Research under Contract No. F44620-70-C-0030 and by CISE Laboratories, Milano, Italy.

†On leave of absence from Institut für Theoretische Physik, Universität Stuttgart, Stuttgart, Germany.

¹R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A **4**, 302 (1971).

²See references in I.

³J. Eberly [Acta Phys. Polon. (to be published)] has recently discussed the case of inhomogeneous broadening.

⁴See, e.g., N. E. Rehler and J. H. Eberly, Phys. Rev. A **3**, 1735 (1971).

⁵The degeneracy quantum number has to be taken into account in order to make $|\alpha r m\rangle$ a complete set of states in the Hilbert space of N two-level atoms.

⁶Off-diagonal elements with respect to α, r do not contribute to any of the expectation values $\langle R^{+l} R^{-m}(t) \rangle$. Off-diagonal elements with respect to m determine the "phased" quantities $\langle R^{+l} R^{-m} \rangle$ for $l \neq m$; their equations of motion can be solved similarly as the above ones for the $p(r, m, t)$; note that $W_{\alpha r m, \alpha r l}(0) = 0$ for $l \neq m$ entails the vanishing of these matrix elements for all times.

⁷Compare I, Sec. IV.

⁸R. M. Dicke, Phys. Rev. **93**, 493 (1954); and in *Proceedings of the Third International Conference on Quantum Electronics, Paris, 1963*, edited by N. Bloembergen

and P. Grivet (Columbia U. P., New York, 1964), Vol. I.

⁹One has to use the commutation relations of the R_i^\pm : $[R_i^+, R_j^+] = 0$ for $i \neq j$; $R_i^+ R_i^- + R_i^- R_i^+ = 1$; $(R_i^\pm)^2 = 0$.

¹⁰Recall that the classical intensity is obtained by neglecting the dispersion of R_3 , i.e., putting $\sum_m m^2 p(r, m) = [\sum_m m p(r, m)]^2$.

¹¹That is, $I(t) = I_{\text{class}}(t)$ to within the accuracy of the numerical evaluation of $I(t)$ ($\sim 0.5\%$).

¹²At the first glance the fact that $\sigma_n^2(t)$ assumes negative values might look strange. However, recall that $\sigma_n^2(t)$ is not the dispersion of a Hermitian operator. We emphasize that our main point is the smallness of $|\sigma_n^2(t)|$. If we had $\sigma_n^2(t) < 0$ and, at the same time $|\sigma_n^2(t)| > 1/N$, we could not, of course, characterize the field as behaving classically.

¹³Recall that I_1 is just the probability per second according to which an isolated excited atom would send a photon into the diffraction solid angle of the end-fire mode.

¹⁴This quantity measures the deviation of the exact theory from the "classical analysis" which suppresses all statistical fluctuations by assuming $\langle a^\dagger R^- \rangle = \langle a^\dagger \rangle \langle R^- \rangle$; see I, Appendix B. By a straightforward extension of the considerations in I, Sec. IV, one readily shows $\langle a^\dagger R^- \rangle = -(ig/\kappa) \langle R^+ R^- \rangle$.

¹⁵R. J. Glauber, Phys. Rev. **130**, 2529; **131**, 2766 (1963); J. R. Senitzky, *ibid.* **111**, 3 (1958).