

SUPERRADIANCE AND ENERGY TRANSFER WITHIN A SYSTEM OF ATOMS^{*}

J. P. STONE, A. NITZAN and J. ROSS

*Department of Chemistry, Massachusetts Institute of Technology,
Cambridge, Mass. 02139, U.S.A.*

Received 10 December 1975

We investigate cooperative effects in energy relaxation and energy transfer for N atoms in a thermal radiation field with superradiance master equations as well as a closed set of coupled moment equations. Both spatially large and spatially small systems are considered. For small systems nonlinear rate equations for the energy are related to the moment equations. Symmetry of the small system to interchanging atoms is used to incorporate off-diagonal solutions of the superradiance master equation in expressions for the probability of the transfer of energy from one group of atoms to another. The long time excitation probability for initially unexcited atoms is large and strongly correlated. Cooperative processes in a large system which fall off with the distance between a cooperating pair of atoms include energy loss and transfer terms in the master equation. The energy transfer is oscillatory in time. Energy relaxation is shown by numerical solution to become cooperative in a very sudden manner as the scale of the atomic system is decreased through the resonant wavelength.

1. Introduction

Superradiance, or collective cooperative emission by a system of atoms or molecules, has received much attention in recent years¹⁻¹⁰). For a system of N two-level atoms located on fixed lattice sites, the appropriate hamiltonian is

$$H = \varepsilon \sum_{j=1}^N \sigma_j^z + \sum_{j=1}^N \sum_k (\sigma_j^+ a_k \exp(ik \cdot x_j) + \sigma_k^- a_k^+ \exp(-ik \cdot x_j)) + H_B, \quad (1.1)$$

where a_k and a_k^+ are operators for the k th field mode in the second quantization representation,

$$H_B = \sum_k a_k^+ a_k$$

^{*} Supported in part by the National Science Foundation and by Project SQUID, Office of Naval Research.

and x_j is the position of the j th atom. The quantities σ_j are operators on the space of the j th two level atom with ground state $|0\rangle$ and excited state $|1\rangle$ defined as

$$\sigma_j^z = \frac{1}{2} |1\rangle \langle 1| - \frac{1}{2} |0\rangle \langle 0|,$$

$$\sigma_j^+ = |1\rangle \langle 0|,$$

$$\sigma_j^- = |0\rangle \langle 1|.$$

We have incorporated the dipole and rotating wave approximations¹¹⁾ in the form of the atom-field interaction.

Dicke first showed¹⁾ that for an atomic system having dimensions much smaller than the resonant wavelength, cooperative spontaneous emission with a maximum rate proportional to N^2 is predicted from a group of atoms if all the atoms are treated as a single quantized system. He defined a cooperativity number r , related to the eigenvalues $r(r+1)$ of the operator R^2 , which is defined [see eqs. (2.1) and (2.6)] in analogy with the operator for the total angular momentum for systems consisting of N spins. The other quantum number m describing the atoms is the eigenvalue of the operator R^z , the z component of the angular momentum in the spin analogy, and corresponds to the degree of excitation of the atoms $m = \frac{1}{2}(n_{\text{exc}} - n_{\text{unexc}})$, where n_{exc} and n_{unexc} are the numbers of excited and unexcited atoms. Dicke's superradiant states are those with large values of r and $m = 0$. Dicke's conclusions concerning the possibility of having emission rates proportional to N^2 were based on first order perturbation theory; however, recent applications of the methods used to study irreversible processes to the problem have confirmed his conclusions. These methods have culminated in the use of master equation techniques²⁾ to provide a complete description of the system of atoms as it approaches equilibrium with the bath-like radiation field. Both approximate and exact analytic solutions of these master equations in the space of Dicke states have been studied. In particular the description of the relaxation by means of nonlinear rate equations for the average energy has been considered¹⁰⁾ as well as the nature of fluctuations from the solutions for the deterministic rate equation⁵⁾. The description of the behavior of systems initially excited locally rather than in a Dicke state is in principle a simple extension of the known description in Dicke states. The only difficulty is that, with certain exceptions, expressions for Dicke states in terms of locally excited states have not been worked out.

Master equations to describe superradiance from spatially large systems have also been derived²⁾, but solutions are lacking. Bonifacio and others⁶⁾ have considered approximate solutions appropriate for a needle-shaped cavity. Also Lee, Lee and Chang⁷⁾ have studied spatially large systems with small numbers of atoms using multiple time scale perturbation theory. Rehler and Eberly⁸⁾ con-

sider the dynamics of large systems with many atoms using a quite different quantum electrodynamic approach.

Some authors consider also processes which compete with superradiant ones, including amplified spontaneous emission^{1,2)} and various propagation effects^{1,3)}. However, we consider here only superradiant processes and therefore in effect assume conditions under which the field functions as a heat bath either at zero (vacuum state field) or some finite temperature.

Superradiant emission arises from cooperative effects among atoms coupled to a common radiation field. In this paper we take into account the initial spatial distribution of energy and extend the analysis of these cooperative effects to include cooperative energy transfer among the atoms. In particular, the transfer of energy from the originally excited atoms to other atoms in the system is considered. In the limit for small systems, atomic systems initially excited in a Dicke state (eigenstate of R^2) with $r < \frac{1}{2}N$, come to a state of final equilibrium with a radiation field at zero-temperature, which is above the ground state energy. The probability distributions in the long time limit, which we find for small systems initially excited locally, show analogous effects, with the added feature that large excitation probabilities are predicted for exactly those atoms which were unexcited initially. We also consider the transfer of energy through the radiation field in the case of atomic systems whose size is comparable to the resonant wavelength in order to begin to assess the effects of spatial separation on the conclusions drawn for the spatially small system.

We begin in section 2 by giving a derivation of a closed set of coupled moment equations for the energy of the atoms by using the cumulant expansion method. Similar moment equations⁵⁾ have appeared previously as moment expansions of the superradiance master equation, but we give a direct derivation which bypasses the master equation and present a new method for closing the equations, which is a generalization of techniques commonly employed for one or two particle systems.

The most elementary indication of the influence which atoms, coupled to a common radiation field, exert on each other is the nonlinear form of rate equations which describe the loss of energy to the radiation field. Such rate equations^{1,0)} are quantitatively correct for early times and describe the qualitative features of superradiant emission from Dicke states. The usual rate equation is obtained from the coupled moment equations by considering only the first equation and replacing $\langle (R^z)^2 \rangle$ in that equation by $\langle R^z \rangle^2$. These equations are assumed to be good approximations when $\langle R^z \rangle \sim N$, where N is the (macroscopically large) number of atoms. Several authors have studied the range of validity of the rate equations⁵⁾. We consider here coupled moment equations for atoms coupled to a radiation field in thermal equilibrium at temperature T as well as the usually considered zero temperature limit (*i.e.*, vacuum state field). In the vacuum case, we show that if the coupled moment equations are modified by retaining in each equation only

those terms $\langle(R^z)^n\rangle$ with the highest exponent n , the solution of the resulting set of equations has the property $\langle(R^z)^n\rangle = \langle R^z \rangle^n$ and therefore corresponds exactly to the nonlinear superradiance rate equation. We show that for a non-zero temperature field this same approximation is justified when the value of the photon number in each resonant mode is small compared to N . In either case, the neglect of terms beyond highest order in $\langle(R^z)^n\rangle$ is not a uniformly good approximation for all the equations, the higher ones of which contain coefficients proportional to powers of N . We consider the effect of this nonuniformity by examining the ratio $\langle(R^z)^n\rangle/\langle(R^z)\rangle^n$ as a function of n as calculated by using the full set of coupled moment equations.

In section 3, we study the relationship between the coupled moment equations and the superradiance master equation. In a related appendix, we apply the cumulant method used in the derivation of the coupled moment equations to a general class of operators and thereby deduce the form of the master equation for a general hamiltonian H of the system. The master equation derived in this way is found to be identical to that obtained with the Zwanzig formalism. Continuing the main development, we present the analytic solutions of the master equation for small systems, including in particular the density matrix elements which are off-diagonal in the cooperation number r and which play a significant role in the description of systems with localized excitation. The cooperation numbers r and r' associated with these off-diagonal density matrix equations are constants of motion and the solutions of the equations are expressed as a product of two factors: one factor depends on r and r' but not on the degree of excitation of the system. This factor determines the maximum decay rate and as we shall see is also responsible for collective oscillations of the rate of internal energy transfer between the atoms. The second factor moderates the decay rate in accordance with the degree of excitation of the atomic system. It consists of a product of normalization factors dependent on r and r' as well as on the initial and final energies multiplied by a complicated time-dependent function, the form of which depends only on the initial and final excitations of the atomic system. Therefore we obtain a separation between effects due to the total excitation of the atoms and those due to their "cooperativity".

In section 4, we turn our attention to describing systems whose initial excitation is localized on specified atoms. Such a description is of most interest in connection with systems which are not restricted in size. However, in order to avail ourselves of analytic techniques, we first consider systems much smaller than the resonant wavelength. For such a system, an adequate description of any localized state $|\Psi\rangle$ in relation to a given initial localized state $|\Psi_0\rangle$ is provided by two parameters m and Q , the first of which is the usual $m = \frac{1}{2}(n_{\text{exc}} - n_{\text{unexc}})$ used to describe the total excitation of the system. The second parameter, Q , the "relatedness", is taken to mean the number of *particular* atoms which differ in excitation in states $|\Psi\rangle$ and $|\Psi_0\rangle$. When $|\Psi_0\rangle$ evolves to $|\Psi\rangle$, Q is related to the amount

of internal energy transfer which occurs. Naturally, for a spatially large system additional vectorial parameters are required to specify fully the relationship between two states $|\Psi\rangle$ and $|\Psi_0\rangle$. For a system of 20 atoms the development of a locally excited system in time is studied in terms of the parameters m and Q as well as the total probability that an atom goes from an excited to unexcited state (or the reverse) in the course of time. Probabilities for events in which excitation is transferred from one location to another within the system show oscillatory behavior due to cooperative frequency shifts. These oscillations are absent, however, in the probability that the system of atoms has a given total excitation, even though this excitation may have been initially localized on a specified set of atoms. For locally excited systems in which more than half the atoms are excited initially, we show that the process in which all the initially unexcited atoms become excited in the course of time is highly favored, resulting in radiation-trapped states analogous to those obtained in the decay of Dicke states with $r < \frac{1}{2}N$. We show that those states in which a large amount of radiation is trapped are characterized by a high degree of correlation between excitation probabilities on the different atoms.

In section 5, we discuss the behavior of locally excited states in systems which are not restricted in spatial extent. The dependence of constants in the master equation on the interatomic distances is evaluated and we developed the matrix form of the master equation for the basis of locally excited states. This form is particularly simple when expressed in terms of the relatedness parameters Q . Numerical solution of the coupled equations for the density matrix elements is used to study energy loss and internal energy transfer for a system of six atoms on fixed lattice sites. The transition to relaxation times indicative of cooperative effects takes place rather suddenly as the size of the system decreases. For systems comparable in size to the resonant wavelength, the dependence of probability distributions on energy and energy transfer numbers (Q) is similar to that for systems of negligible spatial extent. Specific geometric effects play an expectedly secondary role for small N . However, there is reason to believe that geometric effects may be more significant when N is macroscopically large. For example, Bonifacio and Lugiato⁶⁾ have found important directional effects in the net radiation emitted from pencil shaped samples containing large numbers of atoms whose spacing is *large* compared to the resonant wavelength. For energy transfer effects, which are essentially local in nature, this kind of cooperative geometric effect is less likely to be important.

2. Closed coupled moment equations

We derive by means of a truncated cumulant expansion method coupled equations describing the time evolution of the thermally averaged moments of the

operator $R^z = \sum_j \sigma_j^z$ for the total energy of the atoms coupled to a radiation field. The equations are developed for the case where the radiation field is in thermal equilibrium at temperature T , although later we will specialize our results to the zero temperature limit, corresponding to the vacuum state of the field. Closed equations for the first N moments of R^z are obtained in the case of a spatially small system, for which the hamiltonian reduces to the form

$$H = \varepsilon R^z + F^+ R^- + F R^+ + H_B, \quad (2.1)$$

where

$$F = \sum_k a_k, \quad R^z = \sum_{j=1}^N \sigma_j^z, \quad R^+ = \sum_{j=1}^N \sigma_j^+, \quad R^- = \sum_{j=1}^N \sigma_j^-.$$

Since R^z , R^+ , and R^- are sums over atomic Pauli operators, they satisfy the commutation relations

$$\begin{aligned} [R^z, R^+] &= R^+, \\ [R^z, R^-] &= -R^-, \\ [R^+, R^-] &= 2R^z. \end{aligned} \quad (2.2)$$

Following the derivation of Nitzan and Silbey¹⁴) (see also appendix B for more details), we have for any atomic operator θ which commutes with $H_0 = \varepsilon R^z + H_B$ the relations

$$\langle \theta(t) \rangle = \left\langle \exp_0 \left[i \int_0^t V^x(\tau) d\tau \right] \right\rangle \theta(0), \quad (2.3a)$$

$$\frac{d}{dt} \langle \theta(t) \rangle = \left\langle \exp_0 \left[i \int_0^t V^x(\tau) d\tau \right] \right\rangle \sum_{n=2}^{\infty} \frac{1}{n!} K_n^x(t) \theta(0), \quad (2.3b)$$

where

$$V^x A \equiv [V, A],$$

$$V(t) = \exp(iH_0 t) V(0) \exp(-iH_0 t),$$

$$V(0) = F^+ R^- + F R^+,$$

and $K_2^+(t)$ is the n th order cumulant. Only the first nonvanishing term in the cumulant expansion is retained. This has the form¹⁴⁻¹⁵)

$$\frac{1}{2} K_2^x(t) \theta(0) = - \int_0^t \langle [V(\tau), [V(t), \theta(0)]] \rangle d\tau. \quad (2.4)$$

In order to derive a set of coupled equations for the moments of R^z , we evaluate (2.4) for $\theta(0) = (R^z)^n$, $n = 1, \dots, N$. For this purpose, it is convenient to use the relations, valid for any number a ,

$$\begin{aligned} [R^+, (R^z + a)^n] &= R^+ (R^z + a)^n - R^+ (R^z + a + 1)^n, \\ [R^-, (R^z + a)^n] &= R^- (R^z + a)^n - R^- (R^z + a - 1)^n, \end{aligned} \quad (2.5)$$

which may be derived from (2.2) by induction. Also we note that due to the commutation relations (2.2), the operator

$$R^2 \equiv (R^z)^2 + \frac{1}{2}(R^+ R^- + R^- R^+) \quad (2.6)$$

commutes with R^z , R^+ , and R^- . Therefore R^2 is a constant of motion of the complete system as the atoms come to equilibrium with the radiation field. We have then

$$\begin{aligned} -\frac{1}{2}K_2^x(t) (R^z)^n &= \int_0^t \langle [F(\tau) R^+ e^{i\epsilon\tau} + F^+(\tau) R^- e^{-i\epsilon\tau}, \\ &[F(t) R^+ e^{i\epsilon t} + F^+(t) R^- e^{-i\epsilon t}, (R^z)^n]] \rangle d\tau. \end{aligned} \quad (2.7)$$

Since the thermal averages $\langle F(\tau) F(t) \rangle$ and $\langle F^+(\tau) F^+(t) \rangle$ are zero at equilibrium, we drop these terms at the outset to write

$$\begin{aligned} -\frac{1}{2}K_2^x(t) (R^z)^n &= \int_0^t (e^{-i\epsilon(t-\tau)} \langle [F(\tau) R^+, [F^+(t) R^-, (R^z)^n]] \rangle \\ &+ e^{i\epsilon(t-\tau)} \langle [F^+(\tau) R^-, [F(t) R^+, (R^z)^n]] \rangle) d\tau. \end{aligned} \quad (2.8)$$

Now since all the bath operators commute with all the atomic operators and since thermal averaging affects only the bath operators, we obtain

$$\begin{aligned} -\frac{1}{2}K_2^x(t) (R^z)^n &= [R^+, [R^-, (R^z)^n]] \int_0^t e^{-i\epsilon(t-\tau)} \langle F^+(t) F(\tau) \rangle d\tau \\ &+ R^+ [R^-, (R^z)^n] \int_0^t e^{-i\epsilon(t-\tau)} \langle [F(\tau), F^+(t)] \rangle d\tau \\ &+ [R^-, [R^+, (R^z)^n]] \int_0^t e^{i\epsilon(t-\tau)} \langle F^+(\tau) F(t) \rangle d\tau \\ &- [R^+, (R^z)^n] R^- \int_0^t e^{i\epsilon(t-\tau)} \langle [F(t), F^+(\tau)] \rangle d\tau. \end{aligned} \quad (2.9)$$

By applying (2.5) to the atomic operator expressions in (2.9) in such a way as to bring factors involving R^- and R^+ to the left of factors involving R^z , we find that the operators in the first and third terms both equal

$$(R^+R^- + R^-R^+) (R^z)^n - R^+R^- (R^z - 1)^n - R^-R^+ (R^z + 1)^n \quad (2.10a)$$

while $R^+ [R^-, (R^z)^n]$ and $-[R^+, (R^z)^n] R^-$ both equal

$$R^+R^- [(R^z)^n - (R^z - 1)^n]. \quad (2.10b)$$

The definition (2.6) and the commutation relations (2.2) lead to the identities

$$\begin{aligned} R^+R^- &= R^2 - (R^z)^2 + R^z, \\ R^-R^+ &= R^2 - (R^z)^2 - R^z, \end{aligned} \quad (2.11)$$

which are substituted into the expressions (2.10a) and (2.10b) with the result that there appear only R^2 and the powers $(R^z)^n$. Finally, we notice that at equilibrium the two time bath averages depend only on the time difference. As a result we may make the change of variables $\tau' = t - \tau$ or $\tau' = \tau - t$ to combine the sum of the first and third integrals in eq. (2.9) into the single expression

$$C(t) = \int_{-t}^t e^{i\epsilon\tau'} \langle [F^+(0) F(\tau')] \rangle d\tau'. \quad (2.12a)$$

Similarly, the sum of the second and fourth integrals is

$$B(t) = \int_{-t}^t e^{i\epsilon\tau'} \langle [F(\tau'), F^+(0)] \rangle d\tau'. \quad (2.12b)$$

Putting these changes back into (2.9) gives

$$\begin{aligned} -\frac{1}{2}K_2^x(t) (R^z)^n &= C(t) \{ [2R^2 - 2(R^z)^2] (R^z)^n \\ &\quad - [R^2 - (R^z)^2 + R^z] (R^z - 1)^n \\ &\quad - [R^2 - (R^z)^2 - R^z] (R^z + 1)^n \} \\ &\quad + B(t) \{ [R^2 - (R^z)^2 + R^z] [(R^z)^n - (R^z - 1)^n] \}. \end{aligned} \quad (2.13)$$

We notice that since R^2 is a constant of motion, it does not depend on the initial state of the bath for time $t > 0$. Hence

$$\langle R^2 (R^z)^n \rangle = R^2 \langle (R^z)^n \rangle. \quad (2.14)$$

Therefore when we expand the binomials in $-\frac{1}{2}K_z^x (R^z)^n$ as given by (2.13) and apply (2.3) to the resulting expression we obtain

$$\begin{aligned} \frac{d \langle (R^z)^n \rangle}{dt} = & B(t) \left\{ R^2 \sum_{j=0}^{n-1} \binom{n}{j} \langle (R^z)^j \rangle (-1)^{n+j} + n \langle (R^z)^{n+1} \rangle \right. \\ & \left. - \sum_{j=1}^n \binom{n+1}{j-1} \langle (R^z)^j \rangle (-1)^{n+j} \right\} \\ & + C(t) \left\{ R^2 \sum_{j=0}^{n-2} \binom{n}{j} \langle (R^z)^j \rangle [1 + (-1)^{n+j}] \right. \\ & \left. - \sum_{j=1}^n \binom{n+1}{j-1} \langle (R^z)^j \rangle [1 + (-1)^{n+j}] \right\}. \end{aligned} \quad (2.15)$$

The highest moment appearing on the right-hand side is $\langle (R^z)^{n+1} \rangle$. As a result, the equations as they are written are not closed. However, for a system of N atoms, only N moments of the energy distribution are independent. As a result (see appendix A) it is possible to express $\langle (R^z)^{N+1} \rangle$ as a linear combination of all the lower moments as follows

$$\langle (R^z)^{N+1} \rangle = Q_0(N) \langle (R^z)^{N-1} \rangle + Q_2(N) \langle (R^z)^{N-3} \rangle \dots + \begin{Bmatrix} Q_{N-2}(N) \langle R^z \rangle \\ Q_{N-1}(N) \end{Bmatrix}, \quad (2.16)$$

where

$$Q_k(N) = \frac{1}{2^{k+2}} \binom{N+2}{k+3} W_k(N),$$

$$W_0(N) = 1,$$

$$\begin{aligned} W_l(N) = & 1 - \frac{1}{l+2} \sum_{\substack{k=2 \\ k \text{ even}}}^l \left[(N+2) \binom{l+3}{k+1} \right. \\ & \left. - \frac{N+2-k}{k+1} \binom{l+3}{k} \right] W_{k-2}. \end{aligned}$$

It is generally assumed that since the correlation functions in the integrands of (2.12) are composed of a large number of Fourier components of different frequencies, constructive interference will only occur near the origin in time (*i.e.*, the

correlation time is near $\tau = 0$). As a consequence, we replace the limits on the integrals by $t = \infty$. When this is done, it can be shown that¹⁴⁾

$$C(\infty)/B(\infty) = [\exp(\epsilon/kT) - 1]^{-1}.$$

The quantities $\langle (R^z)^n \rangle$ and R^z appearing in eq. (2.15) are operators in the space of the atomic states. However, since $(R^z)^n$ and R^z commute with each other, the equations reduce to equations for numbers – the expectation values of $(R^z)^n$ and R^z – when considered for initial states $|r, m\rangle$ which are eigenstates of both R^z and R^2 . The equations then describe the development in time of the average energy R^z and of the various moments of the energy distribution as atoms initially in the state $|r, m\rangle$ relax to equilibrium with the radiation field. When the field is in a vacuum state with the number of atoms N macroscopically large and r and m both of order N with $m \lesssim r - 5$, the coupled moment equations (2.15), (2.16) may be interpreted for early times ($Bt \ll 1$) as a set of nonlinear rate equations⁵⁾ involving $\langle R^z \rangle$ and higher powers of $\langle R^z \rangle$.

In the present paper, we consider simple properties of the moment equations (2.15) which correlate with this type of behavior and extend the analysis to include thermal fields ($C \neq 0$). We show that when C/B is of the order of the number of atoms, the description based on nonlinear rate equations breaks down even for very early times.

First, in the vacuum limit, when the initial values of R^z and $(R^z)^n$ are $r(r+1)$ and m^n , where $r \sim N$ and $m \sim N$, we can assume for sufficiently small times that $(R^z)^n$ is of order N^n . The terms in the n th coupled equation (2.15) which are highest order in N are just

$$\frac{d \langle (R^z)^n \rangle}{dt} = Bn \{ \langle (R^z)^{n+1} \rangle - R^z \langle (R^z)^{n-1} \rangle \}, \quad (2.17)$$

provided the *coefficients* in the equation are of order less than N . When n itself is of order N , the coefficient of $\langle (R^z)^{n-q} \rangle$ for small q is of order N^{q+2} , so that the approximation (2.17) is no longer valid. If we nevertheless assume (2.17) for all n , we readily find that the rate of change of the product $\langle (R^z)^q \rangle \langle (R^z)^n \rangle$ is given by

$$\begin{aligned} & \frac{d}{dt} (\langle (R^z)^q \rangle \langle (R^z)^n \rangle) \\ &= Bn \langle (R^z)^q \rangle \{ \langle (R^z)^{n+1} \rangle - R^z \langle (R^z)^{n-1} \rangle \} \\ &+ Bq \langle (R^z)^n \rangle \{ \langle (R^z)^{q+1} \rangle - R^z \langle (R^z)^{q-1} \rangle \}. \end{aligned} \quad (2.18)$$

If we now introduce the factorization $\langle (R^z)^k \rangle \langle (R^z)^j \rangle = \langle (R^z)^{j+k} \rangle$ on the right-hand side of (2.18), we obtain

$$\frac{d}{dt} (\langle (R^z)^q \rangle \langle (R^z)^n \rangle) = (n + q) B \{ \langle (R^z)^{q+n+1} \rangle - R^2 \langle (R^z)^{q+n-1} \rangle \}. \quad (2.19)$$

On the other hand, evaluating (2.17) with n replaced by $q + n$ yields again the right-hand side of (2.19) as the time derivative of $\langle (R^z)^{q+n} \rangle$. Thus the expression obtained for the time evolution is independent of how the moment is divided into factors. Since we have, by definition, that $\langle (R^z)^q \rangle \langle (R^z)^n \rangle = \langle (R^z)^{q+n} \rangle$ at $t = 0$, we see that the form of (2.17) is such that this condition is maintained for all times*.

The approximate equations (2.17) are not justified when $m \sim r$ because when $m = r$ their right-hand sides are identically zero, contradicting the assumption that, at least for small n , the terms in (2.15) that were neglected in (2.17) are negligible compared to terms retained.

When $m < r$, we may also consider approximations of the type (2.17) for the case of thermal fields in equilibrium at temperature $T > 0$. In this case $C \neq 0$. Since the highest order terms multiplying C are of order $\langle (R^z)^n \rangle$, for fairly low temperatures where C is small, the approximate equations (2.19) remain unchanged. On the other hand, for temperatures large enough so C/B is of order N , we must include the leading terms multiplying C in (2.17). When this is done, the factorization is no longer consistent. Physically, the condition $C/B = N$ implies that the expectation value of the number of photons in each resonant mode is equal to N .* Therefore, we have the result that if the number of resonant photons is comparable to the number of atoms, the evolution of the system cannot take place in accordance with a simple nonlinear rate equation which ignores the fluctuations of the energy from its average value. At still higher temperatures, B is negligible compared to C , so that nonlinear terms do not play a significant role in the relaxation process. Therefore for sufficiently high temperatures, the atoms decay exponentially to their equilibrium state without cooperative effects.

We now return briefly to the effect of the inapplicability of the approximation (2.17) for n close to N on the arguments we have made. In making the factorization in (2.18), we assume the correctness of the ansatz for expressions with a total power of R^z equal to $q + n + 1$, whereas we end up proving the consistency for

* We may evaluate the possibility of using the factorization ansatz $\langle \theta_1 \theta_2 \rangle = \langle \theta_1 \rangle \langle \theta_2 \rangle$ for any two operators θ_1 and θ_2 by examining the expression $[V(t), [V(\tau), \theta_1 \theta_2]] = [V(t), [V(\tau), \theta_1]] \times \theta_2 + \theta_1 [V(t), [V(\tau), \theta_2]] + [V(\tau), \theta_1] [V(t), \theta_2] + [V(t), \theta_1] [V(\tau), \theta_2]$. The factorization assumption neglects terms arising from $[V(\tau), \theta_1] [V(t), \theta_2] + [V(t), \theta_1] [V(\tau), \theta_2]$.

* For optical frequencies, the temperature necessary to satisfy this condition is much too high to be attained although, in the microwave region, we may satisfy the condition if N itself is not too large.

total powers equal only to $q + n$. Thus, the consistency of factorizing lower powers of R^z depends on the validity of the ansatz for the next higher power and, as we have remarked, when we go to sufficiently high powers (2.17) is no longer a good approximation for (2.15). The one redeeming feature of the situation is the fact that at $t = 0$ the factorization is exact and, as a consequence, we expect the ansatz to be consistent for early times and small n values, while at later times, the error in the higher order equations makes it impossible to prove the ansatz even for the lower moments.

The reasoning outlined in the preceding paragraph leads to some predictions concerning the way in which the quantities $\langle (R^z)^n \rangle$ deviate from $\langle R^z \rangle^n$ as a function of n . When $m < r$, the initial evolution of the lower moments, but not the higher moments, should be consistent with factorization. As a consequence, deviations of $\langle (R^z)^n \rangle$ from $\langle R^z \rangle^n$ should occur earlier for large n . (This suggests

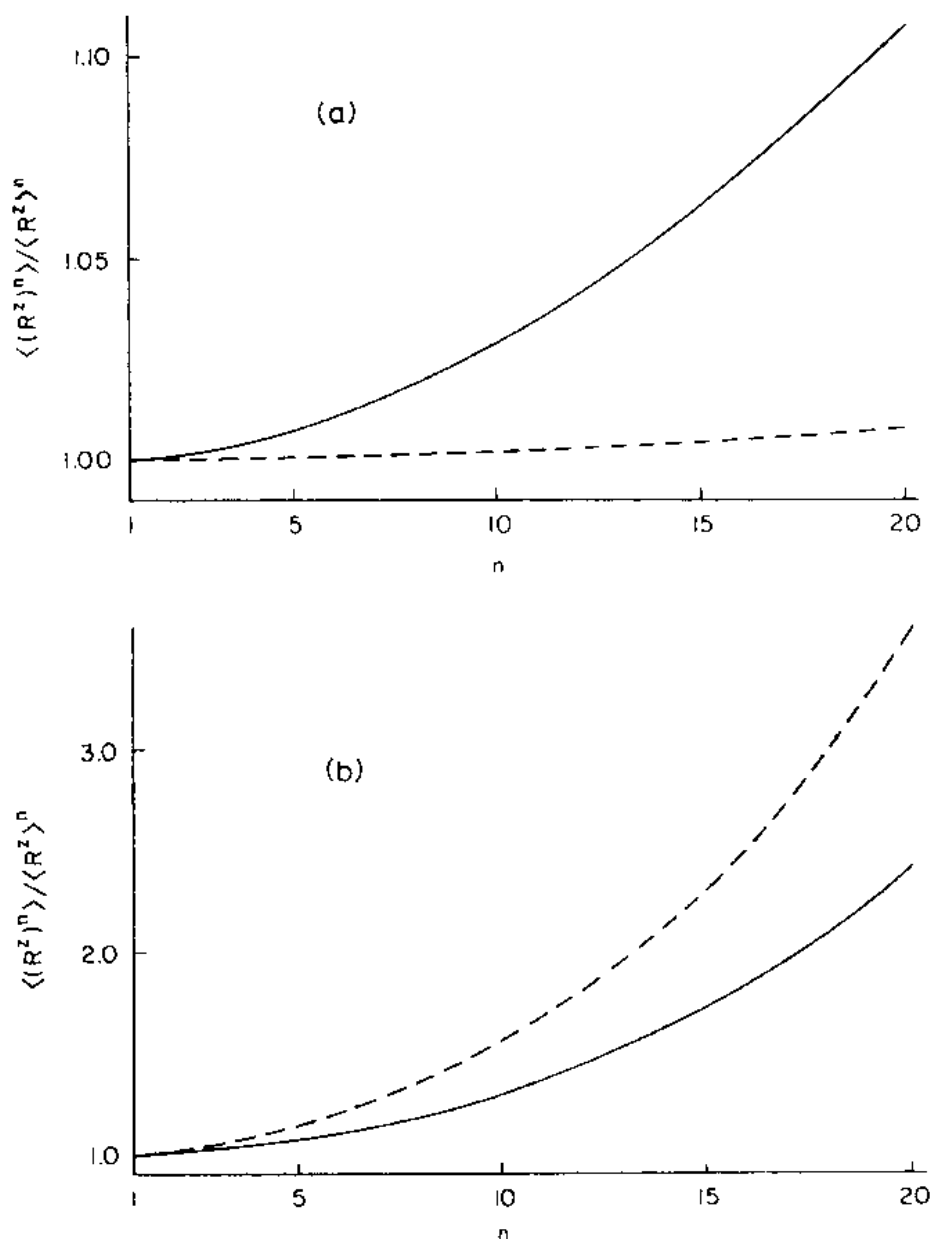


Fig. 1. Deviation of moments from factorized form. (a) $\langle (R^z)^n \rangle / \langle R^z \rangle^n$ as a function of n for $m = 20$ and two values of r at $Bt = 0.0005$, $r = 30$ —, $r = 20$ ---. (b) $\langle (R^z)^n \rangle / \langle R^z \rangle^n$ for the same values of r and m at two later times for which both systems have $\langle R^z \rangle = 18$.

that the usual assumption in truncation of moment equations – that replacement of moments by products of lower order forms is better the higher the moment replaced – must be used with some caution in the present case.) In fig. 1, we have plotted $\langle (R^z)^n \rangle / \langle R^z \rangle^n$ as a function of n for several cases of interest, in each case corresponding to an initial state with $m = 20$. We consider both the case $r = 30$, for which the factorization ansatz should apply, and $r = 20$, for which it should not, and compare these two cases under two sets of circumstances. In *all* cases the deviations increase with n . The first comparison is made for the situation where both systems have been allowed to evolve for a short length of time $t = 0.0005 B^{-1}$ (fig. 1a). The system with $r = 30$ now has $\langle R^z \rangle = 19.72$, whereas the system with $r = 20$ has $\langle R^z \rangle = 19.99$. In this case the deviations for the $r = 30$ case are *larger* because the system has evolved much further from the initial state $\langle R^z \rangle = 20$, for which the factorization assumption is exact. The slow evolution in the case $r = 20$, for which $n = r$, is to be expected in view of the fact that the leading terms in the rate equations are zero in this case. A truer test of our argument is a comparison of the systems at times for which the systems have evolved to the same extent. We have made such a plot in fig. 1b for times when each system has evolved to $\langle R^z \rangle = 18$. In this case the system with $r = 30$ shows, as expected, smaller deviations from the factorized form.

Therefore we see that the trends shown by the deviations from the factorized form are consistent with our method of argument. When the initial conditions are in the factorized form and the lower equations in the coupled set are consistent with maintaining the factorized form, then early in the evolution of the system it is a good approximation to assume the lower moments remain factorized, but it is not such a good approximation to make this assumption for the higher moments.

3. Master equations

The coupled moment equations such as we have discussed in the previous section contain only a limited amount of information about the system. The N moments $(R^z)^n$, $n = 1, \dots, N$ represent N independent linear combinations of diagonal density matrix elements $\rho_{r m \alpha, r m \alpha}$,

$$\langle (R^z)^n \rangle = \sum_{m=-r}^r m^n \rho_{r m \alpha, r m \alpha}, \quad (3.1)$$

and the fact that the equations are closed simply indicates that these diagonal elements are coupled only to each other. Here α is a degeneracy parameter required to specify fully a state with given r and m . Because r and α are both constants of motion, the expression (3.1) for $\langle (R^z)^n \rangle$ does not include sums over r

and α . Similar closed equations for $\langle\langle (R^z)^n (R^+)^n \rangle\rangle$ or $\langle\langle (R^z)^n (R^-)^n \rangle\rangle$ likewise contain information about $\rho_{rm\alpha, rm'\alpha}$ for $m \neq m'$. In fact, by considering a sufficiently general class of operators, complete equations for the density matrix can be generated. Rather than doing this specifically for a superradiant system, in appendix B we derive equations for the density matrix of a more general system by considering coupled thermally averaged operator equations for a complete set of operators. The master equation thus obtained in lowest order cumulant method is identical to that obtained in lowest order by Zwanzig¹⁶).

In the previous section, we have considered coupled moment equations that are valid in the limit of spatially small systems. Since we consider in section 5 systems which are not restricted in size, we include for reference at this point the master equation for the case of hamiltonian (1.1) which is valid regardless of the spatial size of the system. When the prescription for the form of the master equation derived in appendix B is applied to (1.1), the following master equation, similar in general form to one considered by Agarwal²), results

$$\begin{aligned} \frac{d\rho}{dt} = & i\varepsilon \sum_{j=1}^N (\rho\sigma_j^z - \sigma_j^z\rho) \\ & - \sum_{j=1}^N \sum_{i=1}^N [(B_{ij}^- + C_{ij}^-) (\rho\sigma_j^+ \sigma_i^- - \sigma_i^- \rho\sigma_j^+) \\ & + C_{ij}^- (\sigma_i^- \sigma_j^+ \rho - \sigma_j^+ \rho\sigma_i^-) + C_{ji}^+ (\rho\sigma_j^- \sigma_i^+ - \sigma_i^+ \rho\sigma_j^-) \\ & + (B_{ji}^+ + C_{ji}^+) (\sigma_i^+ \sigma_j^- \rho - \sigma_j^- \rho\sigma_i^+)], \end{aligned}$$

where

$$C_{\alpha\beta}^+ = (C_{\alpha\beta}^-)^* = \sum_k \int_0^\infty \exp\{i(\varepsilon - |k|c)\tau\} \langle a_k^+ a_k \rangle \times \exp\{ik \cdot (\mathbf{x}_\beta - \mathbf{x}_\alpha)\} d\tau,$$

$$B_{\alpha\beta}^+ = (B_{\alpha\beta}^-)^* = \sum_k \int_0^\infty \exp\{i(\varepsilon - |k|c)\tau\} \langle [a_k, a_k^+] \rangle \exp\{ik \cdot (\mathbf{x}_\beta - \mathbf{x}_\alpha)\} d\tau.$$

These quantities are related to the constants B and C defined previously in eq. (2.12) by

$$\begin{aligned} (C^-)^* = C^+ &= \lim_{\mathbf{x}_\beta \rightarrow \mathbf{x}_\alpha} C_{\alpha\beta}^+, & C &= C^+ + C^-, \\ (B^-)^* = B^+ &= \lim_{\mathbf{x}_\beta \rightarrow \mathbf{x}_\alpha} B_{\alpha\beta}^+, & B &= B^+ + B^-. \end{aligned} \tag{3.2}$$

Up to this point, we have considered the properties of atomic systems coupled to a field in the vacuum state (zero temperature) as well as systems coupled to thermal radiation fields. However, throughout the remainder of the paper, we devote

our attention to the zero temperature case in which $C_{ji}^+ = C_{ji}^- = 0$. In the remainder of this section and also in the following section, we also consider the limiting case of a spatially small system before returning to the system of unrestricted size in section 5. Thus the master equation which is the basis for our considerations in this and the following section has the form.

$$\frac{d\rho}{dt} = i\varepsilon [\rho, R^z] - B^- \rho R^+ R^- - B^+ R^+ R^- \rho + B R^- \rho R^+. \quad (3.3)$$

By using the relations

$$\begin{aligned} R^z |r, m, \alpha\rangle &= m |r, m, \alpha\rangle, \\ R^+ |r, m, \alpha\rangle &= [r(r+1) - m(m+1)]^{\frac{1}{2}} |r, m+1, \alpha\rangle, \\ R^- |r, m, \alpha\rangle &= [r(r+1) - m(m-1)]^{\frac{1}{2}} |r, m-1, \alpha\rangle, \end{aligned} \quad (3.4)$$

we may write the matrix elements of the master eq. (3.3) in the basis of Dicke states

$$\begin{aligned} \frac{d\rho_{r m \alpha, r' m' \alpha'}}{dt} &= [i\varepsilon (m' - m) - B^- \nu_{m'-1}(r') - B^+ \nu_{m-1}(r)] \\ &\quad \times \rho_{r m \alpha, r' m' \alpha'} + B \nu_m^{\frac{1}{2}}(r) \nu_m^{\frac{1}{2}}(r') \rho_{r m+1 \alpha, r' m'+1 \alpha'}, \end{aligned} \quad (3.5)$$

where

$$\nu_m(r) = r(r+1) - m(m+1).$$

Since coupling is limited to matrix elements having the same energy difference $m - m'$, we can restrict our attention to equations for matrix elements diagonal in the energy. In this case, we write*

$$\begin{aligned} \frac{d\rho_{r m \alpha, r' m \alpha'}}{dt} &= -\text{Re } B^+ ([\nu_{m-1}(r) + \nu_{m-1}(r')] \rho_{r m \alpha, r' m \alpha'} \\ &\quad - i \text{Im } B^+ [r(r+1) - r'(r'+1)] \rho_{r m \alpha, r' m \alpha'}) \\ &\quad + B \nu_m^{\frac{1}{2}}(r) \nu_m^{\frac{1}{2}}(r') \rho_{r m+1 \alpha, r' m+1 \alpha'}. \end{aligned} \quad (3.6)$$

* If we take eq. (3.6) with $r = r', \alpha = \alpha'$, the moments $\langle (R^z)^n \rangle$ can be expressed in terms of the density matrix elements $\rho_{r m \alpha, r m \alpha}$. The solution to eq. (3.6) involves $(2r+1)$ (some degenerate) eigenvalues $\nu_{m-1}(r)$, $-r \leq m \leq r$, which determine a sum of exponential time dependences. On the other hand, we expect on general principles that the solution to (2.15, 16) involves N exponentials. The extra exponentials in eq. (2.15) actually correspond to exponentially increasing terms. However, this does not imply the existence of any instability since the initial conditions for $(R^z)^n$ associated with these exponentially increasing terms cannot be attained for any distribution of energy among the available energy levels - *i.e.*, levels satisfying $-r \leq m \leq r$.

In these equations, m is restricted to be smaller in absolute magnitude than both r and r' . When $m = r(r')$, then $v_m(r)$ ($v_m(r')$) is zero and the implied coupling to an energy state $m + 1$ is not present.

Consider now the solutions to (3.6) for the initial condition $\rho_{rm_1\alpha, r'm_1\alpha'} = 1$, $\rho_{rm_f\alpha, r'm_f\alpha'} = 0$ for $m_f \neq m_1$. Since the field is characterized by zero temperature, the probability of having a final state $m_f > m_1$ is zero at all times. For $m_f \leq m_1$, we write

$$\begin{aligned} \rho_{rm_f\alpha, r'm_f\alpha'} &= \left(\prod_{n=m_f}^{m_1-1} v_n^\dagger(r) v_n^\dagger(r') \right) \exp \left\{ -\frac{1}{2} B [r(r+1) + r'(r'+1)] t \right\} \\ &\quad \times \exp \left\{ -iB' [r(r+1) - r'(r'+1)] t \right\} c_{m_f}(t), \end{aligned} \quad (3.7)$$

where $B' = \text{Im } B^+$. We readily see that $c_m(t)$, as defined by the relationship (3.7), satisfies

$$\frac{dc_m}{dt} = Bm(m-1)c_m(t) + Bc_{m+1}(t) \quad (3.8)$$

subject to the initial condition $c_{m_1}(0) = 1$. The coupled equation (3.8) may be solved by successive application of the familiar formula for the solution of a first order differential equation in a single variable. We express the solutions of (3.6) obtained in this manner in terms of a propagator for the density matrix T , which is a tetradic operator with the property

$$\rho_{ij}(t) = \sum_{kl} T_{ij,kl}(t, 0) \rho_{kl}(0), \quad (3.9)$$

where each index runs over a complete set of quantum states (r, m, α) , $i = \{r(i), m(i), \alpha(i)\}$, etc. The fact that coupling in (3.6) is limited to those matrix elements of ρ , $\rho_{rm\alpha, r'm'\alpha'}$ characterized by the same values of r, r', α, α' , and $m - m' = 0$ is reflected in T as

$$\begin{aligned} T_{ij,kl} &= T_{ij,kl} \delta(\{r(i), r(j), \alpha(i), \alpha(j), m(j) - m(i)\}, \\ &\quad \times \{r(k), r(l), \alpha(k), \alpha(l), m(k) - m(l)\}). \end{aligned} \quad (3.10)$$

We consider now the set of coupled equations (3.6) for particular values of the parameters r, r', α , and α' for the initial condition $\rho_{rm_1\alpha, r'm_1\alpha'} = 1$, $\rho_{rm\alpha, r'm\alpha'} = 0$, $m \neq m_1$. The propagator matrix elements $T_{ij,kl}$ which can be evaluated by considering this particular subset of all the density matrix equations are those for

which

$$\begin{aligned} & \{r(i), r(j), \alpha(i), \alpha(j), m(j) - m(i)\} \\ & = \{r(k), r(l), \alpha(k), \alpha(l), m(k) - m(l)\} = (r, r', \alpha, \alpha', 0), \\ & m(j) = m(i) = m_f, \quad m(k) = m(l) = m_i. \end{aligned} \quad (3.11)$$

Now if we suppress the indices $r, r', \alpha,$ and α' and replace ij and kl by the single indices m_f and m_i , we can write the propagator $T_{m_f m_i}$ as follows:

(1) For $m_i > 0$ and

(a) $m_f > 0,$

$$\begin{aligned} T_{m_f m_i} & = F(m_f, m_i, r, t) F^*(m_f, m_i, r', t) \\ & \times \sum_{l=m_f-1}^{m_i-1} G_1(l, m_f, m_i) \exp\{Bl(l+1)t\}; \end{aligned} \quad (3.12a)$$

(b) $-m_i + 1 \leq m_f \leq 0,$

$$\begin{aligned} T_{m_f m_i} & = F(m_f, m_i, r, t) F^*(m_f, m_i, r', t) \\ & \times \left\{ \sum_{l=0}^{|m_f|} G_3(l, m_f, m_i) \left(Bt + \sum_{n=m_f-1}^{m_i-1} \frac{1}{(n+l)(n+l+1)} \right) \right. \\ & \left. \times \exp\{Bl(l+1)t\} + \sum_{l=|m_f|+1}^{m_i-1} G_1(l, m_f, m_i) \exp\{Bl(l+1)t\} \right\}; \end{aligned} \quad (3.12b)$$

(c) $m_f < -m_i + 1,$

$$\begin{aligned} T_{m_f m_i} & = F(m_f, m_i, r, t) F^*(m_f, m_i, r', t) \\ & \times \left\{ \sum_{l=0}^{m_i-1} G_3(l, m_f, m_i) \left(Bt + \sum_{n=m_f-1}^{m_i-1} \frac{1}{(n+l)(n+l+1)} \right) \right. \\ & \left. \times \exp\{Bl(l+1)t\} + \sum_{l=m_i}^{|m_f|} G_2(l, m_f, m_i) \exp\{Bl(l+1)t\} \right\}. \end{aligned} \quad (3.12c)$$

(2) For $m_i \leq 0,$

$$\begin{aligned} T_{m_f m_i} & = F(m_f, m_i, r, t) F^*(m_f, m_i, r', t) \\ & \times \sum_{l=|m_i|}^{|m_f|} G_2(l, m_f, m_i) \exp\{Bl(l+1)t\}, \end{aligned}$$

where

$$F(m_r, m_i, r, t) = \left(\prod_{n=m_r}^{m_r-1} \nu_n^{\frac{1}{2}}(r) \right) \exp \left\{ -\frac{1}{2} B r (r+1) t \right\} \exp \left\{ i B' r (r+1) t \right\},$$

$$G_1(l, m_r, m_i) = \frac{(2l+1)(l+m_r-1)!(-1)^{m_i+l+1}}{(l-m_r+1)!(l+m_i)!(m_i-l-1)!},$$

$$G_2(l, m_r, m_i) = \frac{(2l+1)(l-m_i)!(-1)^{m_r+l}}{(l-m_r+1)!(l+m_i)!(|m_r|-l)!},$$

$$G_3(l, m_r, m_i) = \frac{(2l+1)^2(-1)^{m_i+m_r+1}}{(l-m_r+1)!(l+m_i)!(m_i-l-1)!(|m_r|-l)!}$$

and \sum'' indicates omission of singular terms. When $r = r'$, the solutions above are equivalent to solutions of the superradiance master equation which have appeared previously¹⁷). We need solutions with $r \neq r'$ in order to analyze the case of locally excited initial states which is the topic of the following section.

The analytical form of the expressions given in (2.12) for $T_{m_r m_i}$ is interesting from the point of view of the separation which occurs between the dependence on the variables r and r' and the variables m_i, m_r . Aside from the time independent factor $\prod_{n=m_r}^{m_r-1} \nu_n^{\frac{1}{2}}(r)$, $T_{m_r m_i}$ is written in a form of a factor which depends only on r and r' and one which depends only on m_r and m_i . The factor which depends on r and r' takes into account the cooperative effects between the atoms, while the other factor gives the dependence of the relaxation rate on the amount of excitation present. The product form is quite important in the problem of reducing the computational effort involved in analyzing locally excited states because it allows us to find $T_{m_r m_i}$ for many different pairs (r, r') by doing a single calculation and multiplying the results by the appropriate exponential factors. This becomes especially important when the number of atoms is large. For $N \gtrsim 25$ the numerical evaluation of the formulas (3.12) is so sensitive to round-off errors that the use of such formulas must be replaced by a numerical integration procedure for the coupled differential equations (3.6). The numerical integration of one set rather than many such sets of coupled equations renders this a workable procedure.

4. Locally excited states in a small system

We now determine how eq. (3.12) derived in the preceding section may be used to describe the behavior of a many atom system with excitation initially localized on specified atoms. We begin first with a system whose dimensions are small compared to the resonant wavelength, which may nevertheless be of macroscopic size.

Whereas the collective modes of excitation are characterized as being eigenstates of the operators R^2 and R^z , we may characterize states of localized excitation as being simultaneous eigenstates of the operators $\sigma_1^z, \sigma_2^z, \dots, \sigma_N^z$. Specification of the eigenvalues of these N operators is adequate to specify completely the system. However, we recall that specifying the eigenvalues of R^2 and R^z leaves the system incompletely determined because for $r < \frac{1}{2}N$, there is in general more than one state with these same eigenvalues. Eq. (3.6) shows that the additional degeneracy parameter λ , which is required to specify completely the state, plays no role at all in the time development of the system. In other words, there are many ways of collectively exciting the system which are equivalent in their decay dynamics.

Our approach here is to find the unitary transformation V between Dicke states and locally excited states. In a system which is small compared to the resonant wavelength of interaction with the radiation field, if a specified group of atoms is excited initially, any of the remaining atoms are equally likely as candidates for becoming excited in the course of time. Therefore any physical quantity which has reference to two different localized states, *e.g.*, an initial state and a final state, may depend on the total energies of these states and on the total number of excitations which must be exchanged to go from one state to the other, but cannot depend on the particular atoms which are involved in the exchange. With this point in mind, we define a quantity $Q(\eta, \xi)$ for two localized states $|\eta\rangle$ and $|\xi\rangle$ to be the number of atoms which differ in their state of excitation. This may be further subdivided into two numbers $q(\eta \leftarrow \xi)$ and $q'(\eta \leftarrow \xi)$ defined for an ordered pair of states (initial and final); q' is the number of individual atoms whose state changes from excited to unexcited and q , the number whose state changes from unexcited to excited. Clearly we have the relations

$$q' + q = Q, \quad (4.1)$$

$$q' - q = m_i - m_f. \quad (4.2)$$

We may think of the quantities q and q' , defined with respect to a particular initial state with energy m_i , as new quantum numbers with which to describe the system. The degeneracy of the state $|q, q'\rangle$ then is found by multiplying two combinatorial factors: (1) The number of ways of picking q atoms to excite out of the original $\frac{1}{2}N - m_i$ unexcited atoms. (2) The number of ways of picking q' atoms out of $\frac{1}{2}N + m_i$ atoms. The degeneracy $g'(q, q')$ of $|q, q'\rangle$ is then given by

$$g'(q, q') = \binom{\frac{1}{2}N - m_i}{q} \binom{\frac{1}{2}N + m_i}{q'}. \quad (4.3)$$

Although our rationale for defining the quantities q and q' is that the dynamics of energy transfer in spatially small systems may be described solely in terms of

them, it is clear that the definitions given above for q and q' are also meaningful for systems with linear dimensions comparable to or larger than the resonant wavelength.

In appendix C, we obtain those properties of the unitary transformation V which are necessary for determining the dynamics of a locally excited system. The resulting expressions, eq. (C.16), unlike the transformation itself, contain no reference to the dynamically irrelevant degeneracy parameter α but depend only on the parameters q , m_i and m_f for the relevant pair of localized states and on r , m_i , and m_f for the contributing Dicke states. We use (C.16) to find the expectation value of the energy of atoms [eq. (C.21)] initially in a locally excited state and to find how the probability density (as determined from ϱ) propagates among different states of local excitation (C.28).

First we consider the time development of the energy of a system initially excited to a localized state state $|l\rangle$ having $\langle R^z(0) \rangle = R^z = m > 0$. We have by appendix (C.21) that the expectation value of $R^z(t)$ is*

$$\langle R^z(t) \rangle = \sum_{r=m}^{\frac{1}{2}N} f(r, m, t) W(r), \quad (4.4)$$

where $f(r, m, t)$ is the expectation value of $R^z(t)$ for a Dicke state and the weighting factors $W(r)$, whose sum is unity, are given by

$$\begin{aligned} W(r) &= \left(\binom{N}{\frac{1}{2}N - m} \right)^{-1} g(r) \\ &= \frac{(\frac{1}{2}N - m)! (\frac{1}{2}N + m)!}{(\frac{1}{2}N - r)! (\frac{1}{2}N + r)!} \frac{2r + 1}{\frac{1}{2}N + r + 1}, \end{aligned} \quad (4.5)$$

where $g(r)$ is the degeneracy of the state $|r, m\rangle$. As was pointed out by Dicke in another context, the weighting factor $W(r)$ for $r = m$ will be quite sizeable when N is large and m is some substantial fraction of N . Thus for m ranging from $\frac{1}{4}N$ to $\frac{1}{2}N$, $W(r = m)$ ranges between approximately $\frac{2}{3}$ and 1. The weighting factor for $f(m + 1, m, t)$ ranges between $\frac{2}{9}$ and 0 for the same range of m . Therefore the significant contributions to $\langle R^z(t) \rangle$ come from at most one or two terms and the emission will closely resemble that of the Dicke state $m = r$. In fig. 2, we compare the decay of a locally excited state with the corresponding Dicke state with

* $\langle R^z(t) \rangle$ could also be found with the coupled moment equations (2.15) by incorporating the matrix expression for R^2 in (C.19). Since R^2 is not diagonal, the full operator nature of the equations must then be taken into account.

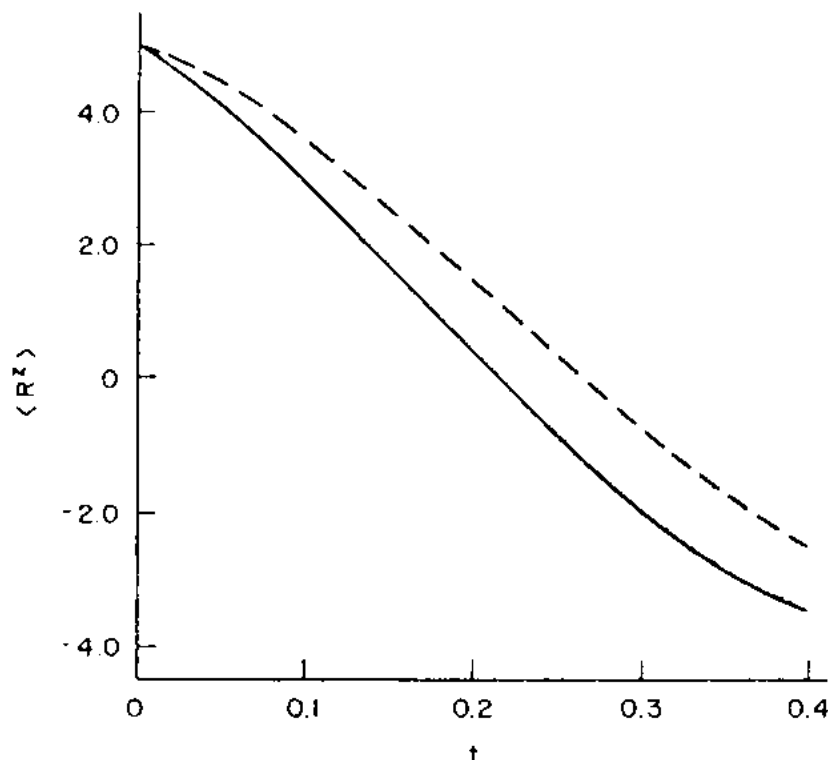


Fig. 2. Decay of atomic excitation in two 20-atom systems with $\langle R^2 \rangle = 5$ initially. (a) ---, Dicke state with $m = 5$, $r = 5$; (b) —, locally excited (uncorrelated) state with 15 particular atoms excited.

$r = m_1$. We note that the effect of terms in (4.4) with $r > m$ is to further steepen a decay curve whose slope is already proportional to N^2 .[‡]

Several authors^{6, 18)} have considered the emission from states which are similar to locally excited states in that the atoms are initially uncorrelated. However they considered only uniformly excited states. Agarwal¹⁸⁾ showed for these states that, unless correlations between the atoms develop, the system must have a dipole moment in order for superradiant emission to occur. On the other hand, for localized states, in which the atoms are also initially uncorrelated and in which the net dipole is zero, we find that superradiant emission is possible. In our system, although the net dipole moment which is proportional to R^+ remains zero at all times, at later times the individual atomic dipoles $\langle \sigma_i^+ \rangle$ do not necessarily retain their initially zero values and, as we shall see, correlations develop between the atoms in the course of time.

[‡] It can be shown that the initial rate of emission as given by (4.2) is proportional to the number, $\frac{1}{2}N + m$, of excited atoms, just as it would be for noncooperative emission. For example, for $m = \frac{1}{2}N - 1$, if we calculate the initial rate of change of $\langle R^2 \rangle$ from $r(r+1) - m(m-1)$, the rate for $|r = \frac{1}{2}N, \frac{1}{2}N - 1\rangle$ is $2(N-1)$, considerably greater than the noncooperative rate, while that for $|r = \frac{1}{2}N - 1, m = \frac{1}{2}N - 1\rangle$ is $N - 2$, slightly less than noncooperative. However, weighting these by the proper degeneracy factors $w(r)$ just gives $N - 1$, exactly the non-cooperative rate. The fact that *initial* rates are proportional to N is, however, in no way inconsistent with obtaining superradiant emission later on, any more than it was for the special case of complete excitation, which is a Dicke state.

We have seen cooperative behavior in the overall decay rate of a locally excited state and now seek to learn if this behavior is also manifest in the rate of intra-system energy transfer. The conditional probability that the system, assumed initially to be in a state of local excitation $|l_0\rangle$, may be found at a later time in a different state of local excitation $|l\rangle$ is given by the matrix element $\mathcal{F}_{ll_0t_0t}$ of the propagator for the density matrix in the basis of localized states which we give below, eq. (4.4). The process of going from $|l_0\rangle$ to $|l\rangle$ involves energy transfer across the system and/or energy loss to the radiation field.

By using the Dicke state propagator developed in (3.12a-d) and the last section of appendix C, we find the following expression for the propagator between localized states

$$\begin{aligned} \mathcal{F}_{ij,kl} &= H(m_i, m_k, t) \\ &\times \left(\sum_{r_u=M}^{\frac{1}{2}N} P_{Q(i,k)}(r_u, m_i, m_k) F(m_i, m_k, r_u, t) \right) \\ &\times \left(\sum_{r_v=M}^{\frac{1}{2}N} P_{Q(j,l)}(r_v, m_l, m_k) F^*(m_l, m_k, r_v, t) \right), \end{aligned} \quad (4.6)$$

where $M = \max\{\text{abs. } m_i, \text{abs. } m_l\}$. Here $H(m_i, m_k, t)$ is defined from eq. (3.12) as

$$H(m_i, m_k, t) = T_{m_i m_k} |F(m_i, m_k, r, t) F^*(m_i, m_k, r', t)|. \quad (4.7)$$

That is, it consists in each case (a-d) of the sums over exponential terms $\exp\{Bl(l+1)t\}$ or $Bt \exp\{Bl(l+1)t\}$. $P_{Q(i,k)}(r, m_i, m_k)$ is a weighting factor specifying the relative importance of contributions from different cooperativity numbers r to the transition moment between two localized states i and k . In appendix (C.1) we have used the symmetry of small systems to the exchange of any two atoms to prove that

$$\begin{aligned} P_Q(r, m', m) &= \left(\frac{(r+m')!(r+m)!}{(r-m')!(r-m)!} \right)^{\frac{1}{2}} \frac{g(r)}{N!} (\frac{1}{2}N - m - q) q'! \\ &\times \sum_{j=0}^q (-1)^j \binom{q}{j} \frac{(r-m+q'-j)!}{(q'-j)!} \frac{(\frac{1}{2}N + m + j - q')!}{(r+m+j-q')!}. \end{aligned} \quad (4.8)$$

Whenever the quantity $B^+ \equiv B + iB'$ defined in (3.2) is complex ($B' \neq 0$), the energy transfer probabilities $\mathcal{F}_{jj,ii}$ obtained from (4.6) together with (3.12) may exhibit oscillatory behavior for $m_i < \frac{1}{2}N$ because of interference effects. These effects will be significant if B' (the Lamb shift) is of the same order as B

(the natural lifetime). A comparison of natural lifetimes and Lamb shifts for typical atomic systems indicates that B' may be of the same order of magnitude as B or larger. As an example, we have calculated the probability $\mathcal{F}_{ll, ll}$ that the system remains in its initial state of local excitation without losing energy to the bath and without transferring excitation to other atoms in the system for systems of 20 and 40 atoms (fig. 3) with $m_l = 5$ and $m_l = 15$, respectively. B' was chosen equal to $8B$. The probability undergoes a single well-defined undulation in each case. Similar undulations are predicted in probabilities for transfer of finite amounts of energy across the system. Since there is no qualitative change in the time dependence of this energy transfer probability in going from 20 to 40 atoms, we surmise that interference oscillations may remain important even for macroscopic atomic systems. We should keep in mind that such undulations have no effect on the observed spontaneous emission since $\langle R^z \rangle$ itself does not depend on oscillating terms [see eq. (4.4)]. Interference effects only become important if an experiment is designed to measure energy transfer across the system.*

Now, we consider several aspects of the behavior of locally excited systems as described by the energy transfer probabilities (4.6). In the ensuing discussions, we assume for simplicity that $B' = 0$.

(a) Evaluating (4.6) with $k = l$ and $i = j$ gives the probability that the system, initially in a pure local state $|l\rangle$ is found at time t in the local state $|j\rangle$, related to $|l\rangle$ through certain energy transfer or energy loss processes. In fig. 4, we plot these probabilities for several values of m_j (and a given initial state with $m_l > m_j$) as a function of the energy transfer number q (the number of initially unexcited atoms which are excited in the final state) for two characteristic times t_1 and t_2 such that $t_1 \ll B^{-1}$ and $t_2 \gg B^{-1}$. At the time t_1 when the system has just begun to relax, we see that the probability for a given value of m_j is a strongly decreasing function of the number of excitations which must be transferred across the system. By the time t_2 when the relaxation process is almost complete, the profile of the system has undergone substantial changes. In the lower energy states, it is now more probable to find the system in a state in which large amounts of excitation have been transferred across the system than in a state in which only small amounts have been transferred. In particular, the most probable state of the system is one which is in a sense the exact opposite of the initial state – exactly those atoms are excited which were not excited initially and all the original excited atoms are in their ground states. However, the asymptotic state reached at the end of the relaxation process is qualitatively different from the initial state in that the density matrix is no longer diagonal (in the local basis). Indeed, the off diagonal density matrix elements must now contain information concerning the mech-

* Bonifacio and Lugiato predict oscillations in the emission from long pencil shaped samples. They point out, however, that these are stimulated emission effects which are specifically excluded from our treatment.

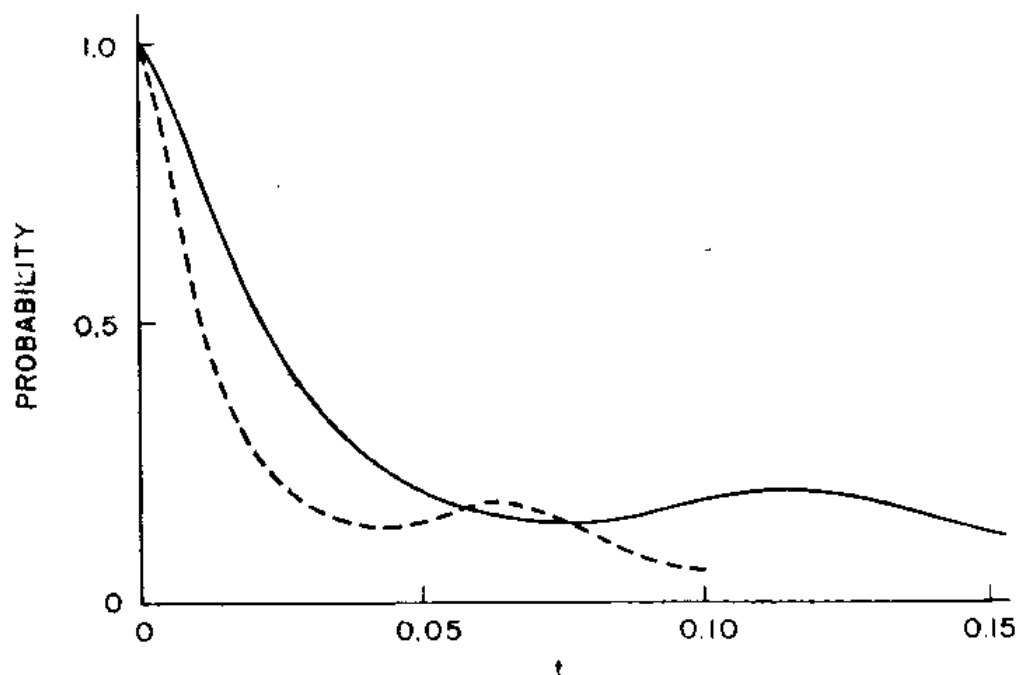


Fig. 3. Interference oscillations in energy transfer probabilities for locally excited systems. Probability that system remains in original locally excited state as function of time. 20 atoms, 15 excited —; 40 atoms, 30 excited ---.

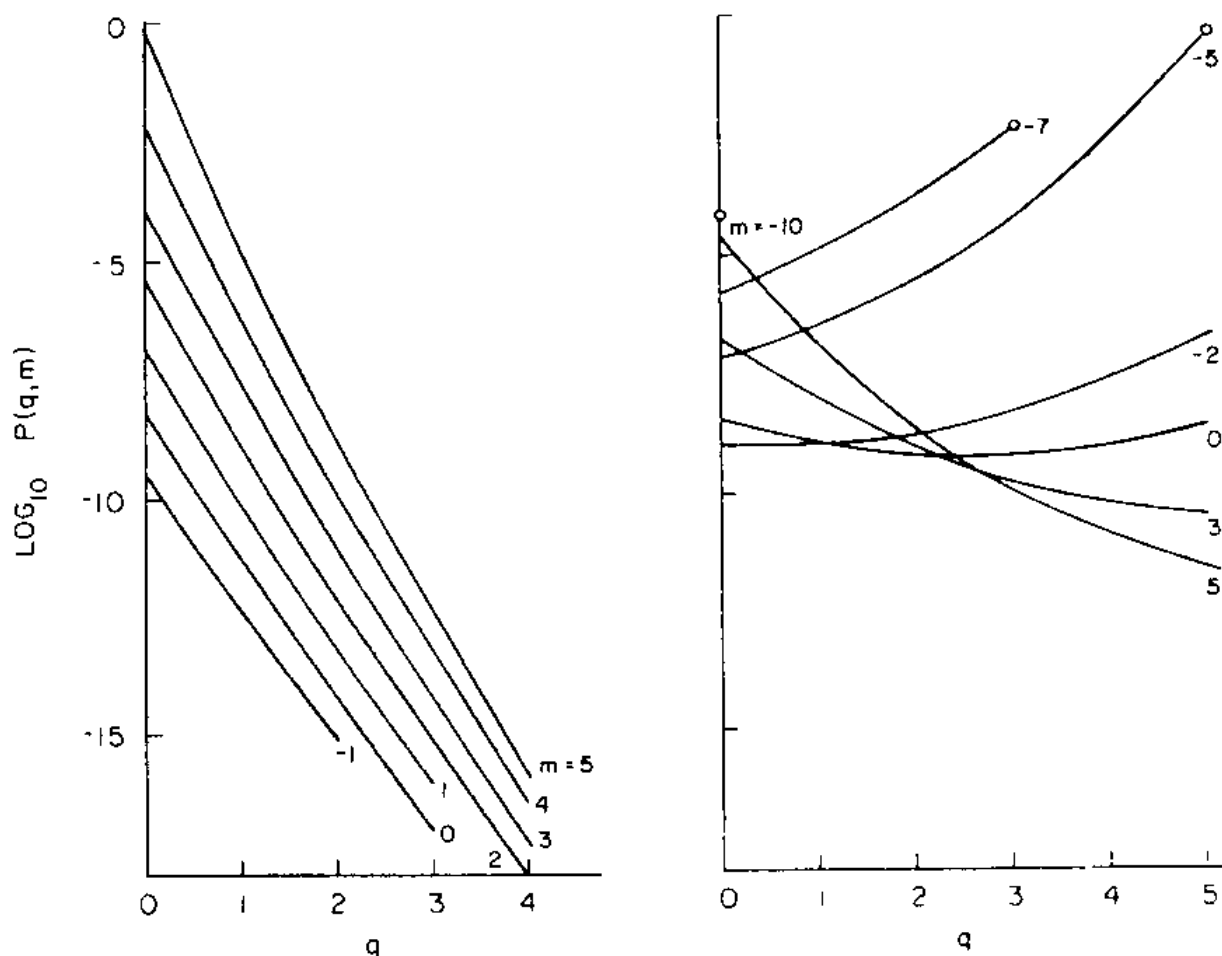


Fig. 4. Probability profile of system as function of energy content and relatedness to initial state. Each curve connects states with same energy m but different values of energy transfer parameter q . System contains 20 atoms, of which 15 are excited initially. (a) Profile for $t \ll B^{-1}$; (b) Profile for $t \gg B^{-1}$.

anism responsible for trapping radiation in the system and preventing its relaxation all the way to the ground state.

(b) The results shown in fig. 4 indicate, at least for one value of N , that in the long time limit, states with large q values are favored. That this is true in general may be seen by looking at expressions for the asymptotic limit of $\mathcal{F}_{VV,II}$ as $t \rightarrow \infty$. Since the strongest dependence on q in fig. 4 was obtained when $m_{i'} = -m_i$, we consider this case also here. Using (4.6) in the limit $t \rightarrow \infty$ together with (C.18), we obtain

$$\rho_{m_i, q, m_i, q} = \left[\frac{(\frac{1}{2}N - m_i - q)! (q + 2m_i)! (2m_i + 1)}{(\frac{1}{2}N + m_i + 1)!} \right]^2 \quad (4.9)$$

which, as expected, has a maximum value for $q = \frac{1}{2}N - m$ given by

$$P = \left(\frac{2m_i + 1}{\frac{1}{2}N + m_i + 1} \right)^2. \quad (4.10)$$

For m_i between $\frac{1}{4}N$ and $\frac{1}{2}N$, P ranges approximately between $\frac{1}{2}$ and 1, which is rather remarkable considering the number of states available to the system. A check may be introduced at this point by noticing that since the asymptotic value of $f(r, m, t)$ in eq. (4.4) is $-r$, the asymptotic energy predicted by (4.4) and (4.6) is the same if

$$\sum_{q=0}^{\frac{1}{2}N-m} \binom{\frac{1}{2}N - m}{q} \binom{\frac{1}{2}N + m}{q'} \rho_{(-m)q, (-m)q} = W(m)$$

for $m \geq m_i$. Here we have introduced the degeneracy factor (4.3). This relationship is verified by (4.9) for the case $m = m_i$.

(c) The probability that an individual atom i in the sample is excited, independently of the states of the other atoms, is obtained from (4.6) by tracing over the states of the remaining atoms and is related to $\langle \sigma_i^z \rangle$ by

$$P_i = \langle \sigma_i^z \rangle + \frac{1}{2}. \quad (4.11)$$

P_i depends only on whether or not i was one of the atoms which was excited originally. In fig. 5 we plot the asymptotic probability that an atom is excited for a 20 atom system as a function of the original degree of excitation of the system. The solid curve gives P_i for the set of atoms i which were initially excited, while the dotted curve gives P_i for atoms which were originally unexcited.

(d) Finally, we consider the role played by correlations between the atoms in our system. Again we consider systems of 20 atoms with varying degrees of initial excitation. This time we start with a state l with $m_l > 0$ and calculate the joint probability \mathcal{P} that all $\frac{1}{2}N - m_l$ atoms which were not excited initially are excited

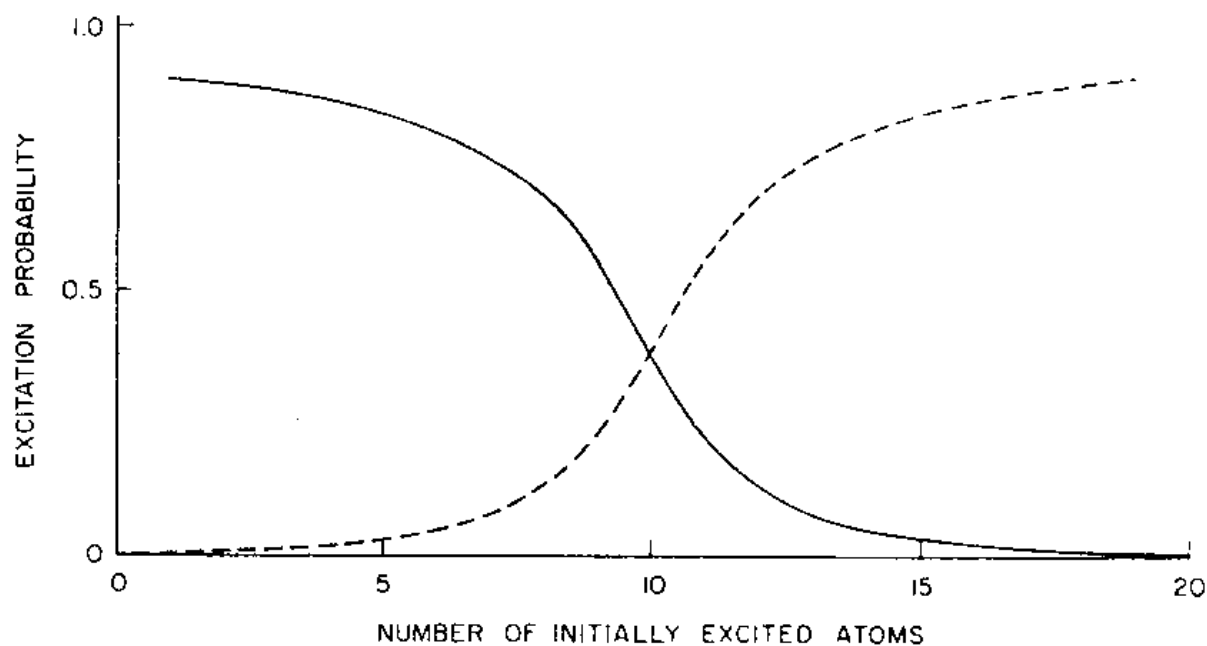


Fig. 5. Long time probability in a locally excited 20-atom system that an atom i is excited as a function of the number $(\frac{1}{2}N - m)$ of atoms excited initially: —, if atom i was excited initially — —, if atom i was unexcited initially.

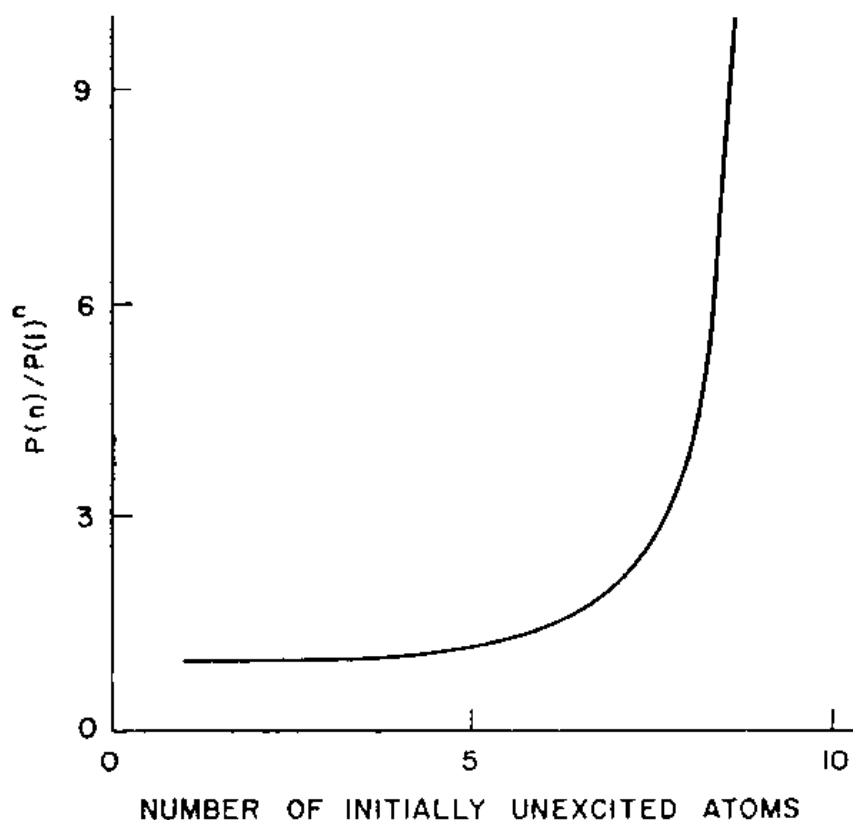


Fig. 6. Correlations in the asymptotic state of a 20-atom system with local excitation; initially n atoms ($n < 10$) are in their ground state. Ratio of probability that all n atoms [eq. (4.7)] are excited to n th power of probability for single atom excitation [dotted line - fig. 5, eq. (4.8)].

asymptotically. Since $-m_i$ is the largest possible value of the asymptotic energy, there is only one quantum state in which a given set of $\frac{1}{2}N - m_i$ atoms is excited, viz., that state $|\Psi\rangle$ in which these atoms are excited and the remaining $\frac{1}{2}N + m_i$ atoms are all unexcited. The probability \mathcal{P} is thus equal to the asymptotic probability of the quantum state $|\Psi\rangle$, i.e., \mathcal{P} is given by eq. (4.10). In the event that the atoms are uncorrelated, we have

$$\mathcal{P}(m_i) = (P_i(m_i))^{\frac{1}{2}N - m_i}, \quad (4.12)$$

where $P_i(m_i)$ is given by (4.11) evaluated for one of the $\frac{1}{2}N - m_i$ initially unexcited atoms i . Therefore, as a measure of the way in which the asymptotic correlations in the system depend on the initial excitation m_i , we plot in fig. 6 the ratio $\mathcal{P}(m_i)/(\mathcal{P}_i(m_i))^{\frac{1}{2}N - m_i}$ as a function of the number of initially unexcited atoms $\frac{1}{2}N - m_i$.^{*} Correlations in the final state become quite large for initial states with a substantial fraction of unexcited atoms. The lack of correlations evident in almost completely excited states in the asymptotic time domain is not representative of the role of correlations at earlier times. Thus, the fact that the Dicke state $r = m = \frac{1}{2}N$, which corresponds to no unexcited atoms in fig. 6, achieves in time an emission rate proportional to N^2 indicates the importance of correlations during the emission process. The atom-atom correlations shown here are associated with the phenomenon of radiation trapping: as the amount of radiation trapped asymptotically increases (which increases with the number of initially unexcited atoms as long as $m_i > 0$), correlations also increase.

5. Local excitation of a large system

In this section, we consider systems of atoms in a radiation field where distances between the atoms are comparable to the resonant wavelength. As that distance becomes large compared to the wavelength, cooperation between the atoms becomes unimportant and only those processes in which each atom loses its energy directly into the radiation field have significant probability.

We again follow the dynamics of the relaxing atoms by considering solutions of the master eq. (3.1), this time without the simplifying assumption that all the interatomic distances $|x_\beta - x_\alpha|$ are negligible. As the first step, we evaluate the dependence of the constants $C_{\alpha\beta}^+$ and $B_{\alpha\beta}^+$ [eq. (3.1)] on the interatomic distances. Because we are considering a zero-temperature radiation field, $C_{\alpha\beta}^+$ vanishes.

The constant $B_{\alpha\beta}^+$ defined in (3.1) is complex. For the spatially small system, we have demonstrated that the effect of the imaginary part is to introduce cooperative level shifts which give rise to undulatory behavior in the time dependence of the

^{*} If $m_i < 0$, the state $|\Psi\rangle$ defined above has more energy than the initial state. Hence $\mathcal{P} = 0$. For this reason we always consider systems in which at least half the atoms are excited.

probability for local excitation. However, we wish to avoid this complication in the present discussion and therefore set $\text{Im } B_{\alpha\beta}^+ = 0$. Noting that $B_{\alpha\beta} = 2 \text{Re } B_{\alpha\beta}^+$, we extend the limits of integration over τ in eq. (3.1), replace the discrete sum over k by an integral, and perform the angular integration to obtain

$$B_{\alpha\beta} = 2\pi \int_0^\infty \langle [a_k, a_k^+] \rangle \frac{\sin k |x_\beta - x_\alpha|}{k |x_\beta - x_\alpha|} \left[\int_{\infty^-}^\infty e^{i(\varepsilon - kc)\tau} d\tau \right] dk. \quad (5.1)$$

By noting that the time integral is just the delta function $\delta(\varepsilon - kc)$, we can finally write

$$B_{\alpha\beta} = B \frac{\sin k_0 |x_\beta - x_\alpha|}{k_0 |x_\beta - x_\alpha|}, \quad \alpha \neq \beta, \quad (5.2)$$

$$B_{\alpha\alpha} = B,$$

where B is the value of the constant for interatomic distances small compared to the resonant wavelength and k_0 is the resonant wave vector*.

The operator form of the master eq. (3.1) is considerably simpler in appearance in the vacuum limit, which we are considering:

$$\frac{d\rho}{dt} = i\varepsilon \sum_{j=1}^N (\rho\sigma_j^z - \sigma_j^z\rho) - \sum_{j=1}^N \sum_{l=1}^N \frac{1}{2} B_{lj} (\rho\sigma_j^+ \sigma_l^- - 2\sigma_j^- \rho\sigma_l^+ + \sigma_j^+ \sigma_l^- \rho). \quad (5.3)$$

Besides setting $C_{ij}^+ = C_{ij}^- = 0$, we have also assumed that B is real and set $B_{ij}^+ = B_{ij}^- = \frac{1}{2} B_{ij}$.

We determine the form of the desired master equation by evaluating the matrix elements of the operators appearing on the right-hand side of (5.3) between two locally excited states α and β . The result is

$$\begin{aligned} \left\langle \alpha \left| \frac{d\rho}{dt} \right| \beta \right\rangle &= i\varepsilon (m_\beta - m_\alpha) \langle \alpha | \rho | \beta \rangle \\ &- \sum_{l=1}^N \sum_{\substack{j=1 \\ s_j(\beta(-l)) < 0}}^N \frac{1}{2} B_{lj} \langle \alpha | \rho | [\beta(-l)] (+j) \rangle \\ &- \sum_{j=1}^N \sum_{\substack{l=1 \\ s_l(\alpha(-j)) < 0}}^N \frac{1}{2} B_{lj} \langle [\alpha(-j)] (+l) | \rho | \beta \rangle \\ &+ \sum_{j=1}^N \sum_{\substack{l=1 \\ s_l(\beta) < 0}}^N B_{lj} \langle \alpha(+j) | \rho | \beta(+l) \rangle. \end{aligned} \quad (5.4)$$

* Agarwal²⁾ has also derived forms for the constants in the large system master equation. However, his hamiltonian differs from (2.1) in that his interaction between an atom and a mode of the light field is a function of the angle between k and the (fixed) direction of the atomic dipole transition moment.

Here $s_j(\alpha) = \langle \alpha | \sigma_j^z | \alpha \rangle$ and the states $|\alpha(+i)\rangle$ and $|\alpha(-j)\rangle$ are defined, when $s_i(\alpha) < 0$ and $s_j(\alpha) > 0$ respectively, as

$$|\alpha(+i)\rangle = \sigma_i^+ |\alpha\rangle, \quad |\alpha(-j)\rangle = \sigma_j^- |\alpha\rangle. \quad (5.5)$$

Otherwise they are undefined. As with the coupled equations (3.5), the energy difference $m_\beta - m_\alpha$ associated with each density matrix element $\rho_{\alpha\beta}$ appearing in a closed subset of the equations is a constant for the entire subset. Since we are interested in diagonal density matrix elements, we consider those equations for which $m_\beta = m_\alpha$.

For computational purposes we characterize the states appearing on the right-hand side of (5.4) in terms of the energy transfer parameters q and q' defined in the beginning of section 4. With the notation $m \equiv m_\alpha = m_\beta$, we find that the energies associated with states in the first two double-sum terms are all equal to m . On the other hand, in the last term $|\alpha(+j)\rangle$ and $|\beta(+l)\rangle$ are both of energy $m+1$. If we consider $|\alpha\rangle$ and $|\beta\rangle$ to be final states, then the respective energy transfer parameters relating them to $|\alpha(+j)\rangle$ and $|\beta(+l)\rangle$ are

$$q'(\alpha \leftarrow \alpha(+j)) = 1, \quad q(\alpha \leftarrow \alpha(+j)) = 0,$$

$$q'(\beta \leftarrow \beta(+l)) = 1, \quad q(\beta \leftarrow \beta(+l)) = 0.$$

In fact, the last term consists of the sum

$$\sum_{\alpha', \beta'} B_{K(\alpha, \alpha'), K(\beta, \beta')} \rho_{\alpha' \beta'}$$

over all those states α' and β' whose energy transfer parameters are

$$\begin{aligned} q'(\alpha \leftarrow \alpha') &= 1, & q(\alpha \leftarrow \alpha') &= 0, \\ q'(\beta \leftarrow \beta') &= 1, & q(\beta \leftarrow \beta') &= 0. \end{aligned} \quad (5.6)$$

The quantity $K(\alpha, \alpha')$ which is defined for any pair of states α and α' satisfying (5.6) is taken to be the label of the atom which changes excitation.

Next we consider the energy transfer parameters associated with terms in the first two double sums. One readily verifies that the conditions on the inner sums

$$s_j(\beta(-l)) < 0 \quad \text{and} \quad s_i(\alpha(-j)) < 0$$

are equivalent to the compound conditions

$$(s_j(\beta) < 0 \quad \text{or} \quad j = l) \Leftrightarrow s_i(\beta(-l)) < 0$$

and

$$(s_l(\alpha) < 0 \quad \text{or} \quad l = j) \Leftrightarrow s_l(\alpha(-j)) < 0$$

under the restriction already imposed in the outer sums. We first consider the terms satisfying $j = l$. In this case $|\{\beta(-l)\}(+j)\rangle = |\beta\rangle$, $\langle\{\alpha(-j)\}(+l)| = \langle\alpha|$ and the heat bath constants (eq. (5.2)) all satisfy $B_{jl} = B_{jj} = B$. Consequently all such terms may be combined. Since there are $\frac{1}{2}N + m$ terms with $l = j$ in each sum, the total contribution is $B(\frac{1}{2}N + m)\varrho_{\alpha\beta}$. Finally, we consider terms with $j \neq l$. The first double sum consists of a sum over all states β' such that

$$q'(\beta \leftarrow \beta') = 1, \quad q(\beta \leftarrow \beta') = 1, \tag{5.7}$$

$$\sum_{\beta'} \frac{1}{2} B_{K_1'(\beta, \beta'), K_2'(\beta, \beta')} \varrho_{\alpha\beta'}$$

Here $K'(\beta, \beta')$ is a two-dimensional vector defined for any pair of states β and β' satisfying (5.7). The component K_1' is the label of the atom which becomes excited in the transition $\beta \leftarrow \beta'$, whereas K_2' labels the atom which becomes de-excited. The analysis for the remaining sum is the same.

We can now rewrite the master eq. (5.4) entirely in terms of the energy transfer parameters:

$$\begin{aligned} \frac{d\varrho_{\alpha\beta}}{dt} = & -(\frac{1}{2}N + m) B \varrho_{\alpha\beta} + \sum_{\substack{\alpha' \\ C(\alpha \leftarrow \alpha')}} \sum_{\substack{\beta' \\ C(\beta \leftarrow \beta')}} B_{K(\alpha, \alpha') K_1(\beta, \beta')} \varrho_{\alpha'\beta'} \\ & - \frac{1}{2} \sum_{\substack{\beta' \\ C'(\beta \leftarrow \beta')}} B_{K_1'(\beta, \beta') K_2'(\beta, \beta')} \varrho_{\alpha\beta'} - \frac{1}{2} \sum_{\substack{\alpha' \\ C'(\alpha \leftarrow \alpha')}} B_{K_1'(\alpha, \alpha') K_2'(\alpha, \alpha')} \varrho_{\alpha'\beta}. \end{aligned} \tag{5.8}$$

Here C and C' denote the following conditions on a pair of states ν and η :

$$\begin{aligned} C(\nu \leftarrow \eta) & \Rightarrow q(\nu \leftarrow \eta) = 0 \quad \text{and} \quad q'(\nu \leftarrow \eta) = 1, \\ C'(\nu \leftarrow \eta) & \Rightarrow q(\nu \leftarrow \eta) = 1 \quad \text{and} \quad q'(\nu \leftarrow \eta) = 1. \end{aligned} \tag{5.9}$$

K and K' are respectively one and two dimensional vectors defined for pairs of states satisfying C and C' respectively. The components of the vectors are the labels of those atoms K for which $s_K(\nu) \neq s_K(\eta)$ i.e., of those atoms differing in excitation in ν and η . (The ordering of the components of K' is immaterial for the present application.)

Thus we see that the rate of change of $\varrho_{\alpha\beta}$ depends on (1) a noncooperative decay rate $-(\frac{1}{2}N + m) B$ proportional to $\varrho_{\alpha\beta}$ itself, (2) terms involving two higher energy states α' and β' which can form α and β through loss of a single excitation

to the radiation field, and (3) terms involving an unchanged state α (or β) and a second modified state β' (or α') of the same energy related to the original state β (or α) through the transfer of a single excitation across the system between two atoms.

Terms in the master eq. (5.8) which involve only a single atom include the decay term $-(\frac{1}{2}N + m) B_{\alpha\beta}$ as well as any terms in the double sum over α' and β' for which $K(\alpha, \alpha') = K(\beta, \beta')$. All remaining terms refer to two atoms j and l and are proportional to the heat bath constant B_{jl} defined in (4.2). Since B_{jl} becomes small when the distance between atoms j and l is large compared to the resonant wavelength λ , only terms involving a single atom are important for systems with intermolecular distances *very* large compared to λ . Thus the cooperative relaxation goes over in this limit to the rate $-(\frac{1}{2}N + m) B$ which characterizes noncooperative relaxation.

The master eq. (5.8) describes two types of processes which are cooperative in nature and which depend, for their existence, on having the atoms sufficiently close that the heat bath constants are not too small. The last two summations describe energy transfer through the field between two atoms K'_1 and K'_2 . On the

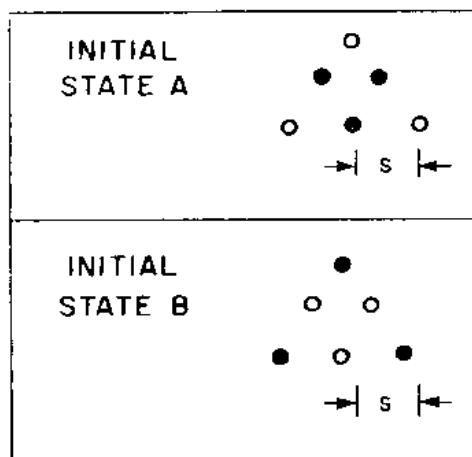


Fig. 7. Two initial localized states used in calculations \bullet indicate excited atom; \circ , ground state atom. Indicated size parameter s may be larger or the same order as $1/k_0$ [eq. (5.2)]; $s \ll 1/k_0$ corresponds to results of section 4.

other hand, the terms of the double sum for which $K(\alpha, \alpha') \neq K(\beta, \beta')$ describes a different type of cooperative process in which two different atoms lose energy to the radiation field. Even though this process can be described in terms of two separate interactions with the radiation field its contribution to the dynamics falls off with the distance R separating the two atoms $K(\alpha, \alpha')$ and $K(\beta, \beta')$ as $\sin kR/kR$ [see eq. (5.2)]. Hence these apparently distinct processes are actually correlated if the atoms are close to each other.

We have studied the solutions to the master eq. (5.8) by means of numerical integration. The closed set of equations including the diagonal density matrix elements contains altogether $\binom{2N}{N}$ coupled equations for a system of N atoms.

We have found it advantageous to construct in advance (*i.e.*, before the point by point integration) tables for each state ν of the states η satisfying $C(\nu \leftarrow \eta)$ and $C'(\nu \leftarrow \eta)$ [eq. (5.9)]. The extremely strong dependence of the number of coupled equations on N has nevertheless restricted our calculations to very small values of N .

For our calculations, we have chosen a collection of six atoms arranged on a regular triangular lattice as shown in fig. 7. Although $N = 6$ is hardly macroscopically large, we expect that some of the behavior observed here, when stated in appropriately general terms, will carry over to larger systems. We consider two initial states, which we designate A and B, which we may characterize in terms of the compactness of the pattern of excitation. In the more compact state A (see fig. 7) the three middle atoms are excited, whereas in the state B, the three outer atoms are excited.

Systems in which interatomic distances are negligible compared to the resonant wavelength have states which correspond to radiation trapping and hence require an infinite amount of time for all the atoms to reach the ground state. In systems where typical interatomic distances s (see fig. 7) are small, but not negligible, the time required for any given large fraction of the atoms to reach the ground state becomes very large as s becomes of the order of the resonant wavelength or smaller. This is illustrated in fig. 8 for initial states A and B. When s is much larger than $1/k_0$, the system decays to the ground state much more rapidly than when $s = 1/k_0$. The decay of states A and B is quite similar even when s equals the resonant wavelength (fig. 8a). In fig. 8b we plot the time required to go from the initial state (in which half the atoms are in their ground states) to the situation where 90% of the atoms are in the ground state as a function of the distance s . This relaxation time is almost constant for $s > 1/k_0$, but begins to increase very rapidly as s is further decreased when $s \approx 1/k_0$. Thus, we obtain a rather sharp transition between regions of cooperative and noncooperative behavior.

We have also made calculations of the cooperative internal transfer of energy for these same states of the six atom system when $k_0s = 1$. The analysis of such results is complicated by the fact that in the spatially extended system, the previously defined energy transfer numbers q' and q are not adequate to define the final state of the system. Nevertheless, if we consider the probability of reaching various final states with given values of q and q' , averaged over all states corresponding to these values, the resulting distributions are qualitatively similar to those obtained for a spatially small system (fig. 4) and thus show the same cooperative energy transfer effects. (In making this comparison we must take $Bt \sim 1$ rather than $t \gg B^{-1}$ for the extended system since when $t \gg B^{-1}$, energy that has at one time been internally transferred will have subsequently been lost to the radiation field.)

Internal transfer of energy should be favored more for an initial state, such as A, with a compact pattern of excitation than for a state B, whose excitation is located

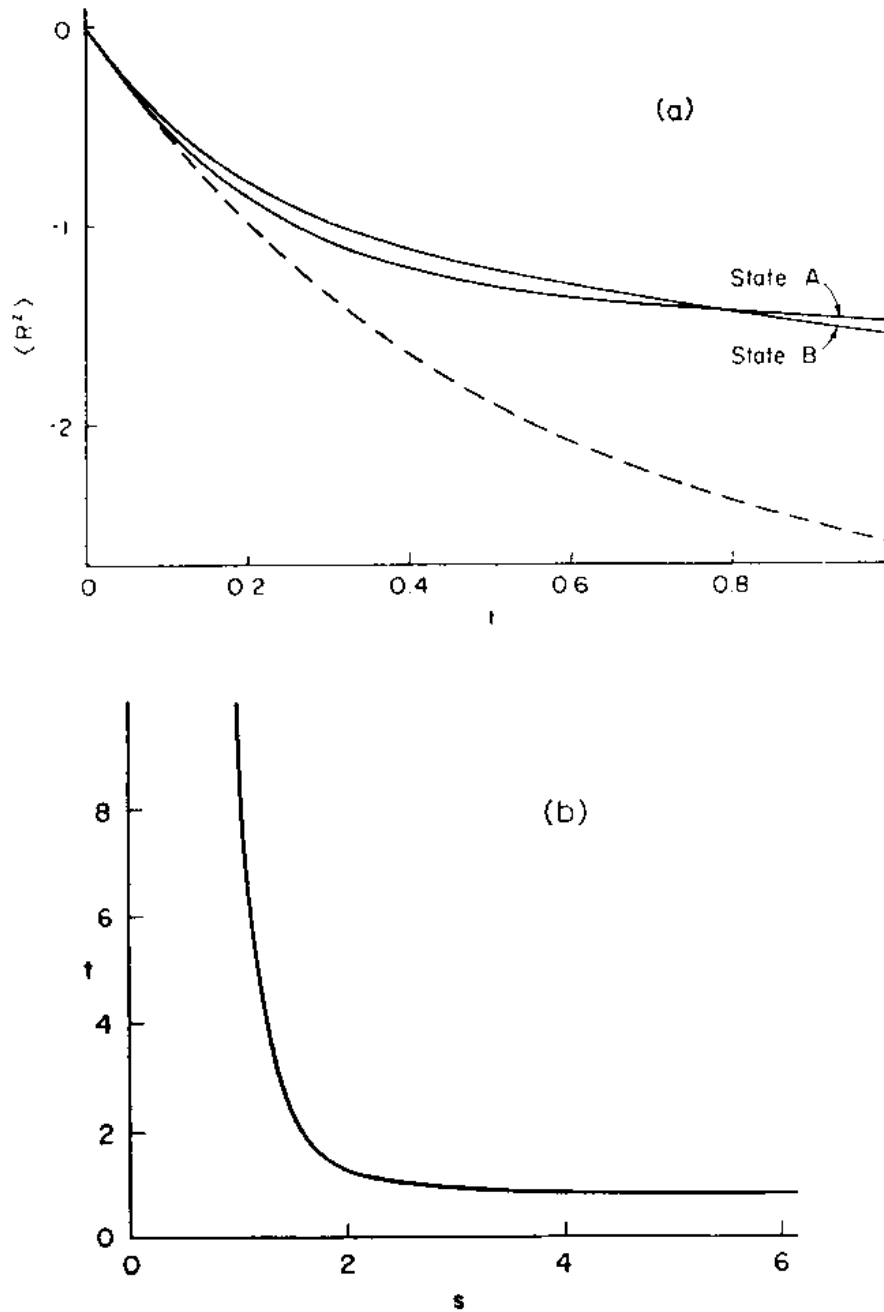


Fig. 8. Energy loss. (a) Decay of total energy in the atoms for the two initial states defined in fig. 7 for $s = 1/k_0$ (solid lines) and $s = 16/k_0$ (dotted line). $\langle R^2 \rangle$ as a function of time. (b) Decay time from initial state B: time required for system to reach $\langle R^2 \rangle = -2.4$ (90% of atoms in ground state) as a function of the distance s . The quantity t in units of B^{-1} , s in units of k_0^{-1} .

near the edges of the system. We find this to be the case, but the effect is relatively small due to the small number of atoms involved.

Acknowledgement

We thank Professor Robert J. Silbey for some helpful discussions concerning the application of the cumulant method.

Appendix A

We show here how to find an expression for the N atom operator $(R^z)^{N+1}$ in terms of lower moments of R^z . We define an $N - 1$ atom operator

$$S^z = \sum_{j=1}^{N-1} \sigma_j^z \quad (\text{A.1})$$

and an N atom operator

$$R^z = \sum_{j=1}^N \sigma_j^z. \quad (\text{A.2})$$

We show by induction that $(S^z)^N$ can be expressed as

$$(S^z)^N = P_{N-2}(S^z), \quad (\text{A.3})$$

where P_{N-2} is a polynomial of degree $N - 2$. This is certainly true for $N - 1 = 1$ since

$$(\sigma^z)^2 = \frac{1}{4}. \quad (\text{A.4})$$

Now expand

$$(R^z)^{N+1} = (S^z + \sigma_N^z)^{N+1},$$

$$(R^z)^{N+1} = (S^z)^{N+1} + (N+1)(S^z)^N \sigma_N^z + \sum_{k=2}^{N+1} \binom{N+1}{k} (S^z)^{N+1-k} (\sigma_N^z)^k \quad (\text{A.5})$$

$$= [S^z + (N+1)\sigma_N^z] P_{N-2}(S^z) + \sum_{k=2}^{N+1} \binom{N+1}{k} (S^z)^{N+1-k} (\sigma_N^z)^k \quad (\text{A.6})$$

$$= [R^z + N\sigma_N^z] P_{N-2}(R^z - \sigma_N^z) + \sum_{k=2}^{N+1} \binom{N+1}{k} (R^z - \sigma_N^z)^{N+1-k} (\sigma_N^z)^k. \quad (\text{A.7})$$

In this expression, even powers of σ_N^z become numerical factors and odd powers become numerical multiples of σ_N^z . R^z is symmetrical to the interchange of any two atoms, say j and N , so we have

$$(R^z)^{N+1} = f(R^z) + \sigma_N^z g(R^z). \quad (\text{A.8})$$

Interchanging atoms j and N , we have

$$(R^z)^{N+1} = f(R^z) + \sigma_j^z g(R^z). \quad (\text{A.9})$$

Since $\sigma_j^z \neq \sigma_N^z$, the expressions (A.8) and (A.9) are consistent only if $g(R^z) = 0$. Therefore we see that $(R^z)^{N+1}$ can be expressed as a polynomial of degree $N - 1$ in R^z . Furthermore, we also see that the only powers of R^z which occur are $(R^z)^{N-1-2k}$, $k = 0, 1, \dots$. So we write

$$P_{N-1}(R^z) = Q_0(N) (R^z)^{N-1} + Q_2(N) (R^z)^{N-3} + \dots \quad (\text{A.10})$$

Having established that this form is correct for all N , we proceed to evaluate the coefficients. Substituting

$$P_{N-2}(R^z) = Q_0(N-1) (R^z)^{N-2} + Q_2(N-1) (R^z)^{N-4} + \dots \quad (\text{A.11})$$

into (A.7) we find an expression for $g(R^z)$ in terms of Q_0, Q_2, \dots . Since each power of R^z must vanish in $g(R^z)$, this gives us a set of conditions that the Q_j 's must satisfy. The result is an explicit functional form for $Q_0(N)$ as a function of N and for recurrence relations that allow functional forms to be found for $Q_2(N), Q_4(N), \dots, Q_{2k}(N), \dots$

$$Q_k(N) = \frac{1}{2^{k+2}} \binom{N+2}{k+3} W_k,$$

where $W_0 = 1$, and W_k satisfies the recurrence relations

$$W_l = 1 - \frac{1}{l+2} \sum_{\substack{k=2 \\ k \text{ even}}}^l \left[(N+2) \binom{l+3}{k+1} - \frac{N+2-k}{k+1} \binom{l+3}{k} \right] W_{k-2}.$$

Appendix B

We derive here equations for the reduced density operator using the cumulant expansion - coupled operator approach and compare them to the corresponding results obtained by Zwanzig's projection operator techniques. We consider a system which consists of three parts - a system of interest, a heat bath, and coupling between the system and the bath

$$H = H_0 + V, \quad H_0 = H_S + H_B. \quad (\text{B.1})$$

We assume that V has the form

$$V = \sum_k B_k S_k, \quad (\text{B.2})$$

where B_i and S_i represent bath and system operators respectively. We further assume that the time dependence of the operators $S_i(t) = \exp(iH_S t) S_i(0) \times \exp(-iH_S t)$ is known and may be expressed in the form

$$S_k(t) = \sum_l \mathcal{S}_{kl} \exp(i\omega_l t). \quad (\text{B.3})$$

For simplicity, we also require that the system be spanned by a denumerable basis $\langle i|$ and from this basis we define a set of operators $\theta_{ij} = |i\rangle \langle j|$. It is convenient to take the basis vectors to be energy eigenstates of H_0 . The time dependence of θ_{ij} is given by

$$\begin{aligned} \theta_{ij}(t) &= \exp(iH^x t) \theta_{ij} \\ &= \exp\left(i \int_0^t V^x(\tau) d\tau\right) \exp(iH_0^x t) \theta_{ij} \\ &= \exp\left(i \int_0^t V^x(\tau) d\tau\right) \theta_{ij} \exp\{i(\epsilon_i - \epsilon_j)t\}, \end{aligned} \quad (\text{B.5})$$

where

$$\begin{aligned} \epsilon_k &= (H_S)_{kk}, \\ \langle \theta_{ij}(t) \rangle &= \exp\{i(\epsilon_i - \epsilon_j)t\} \left\langle \exp\left[i \int_0^t V^x(\tau) d\tau\right] \right\rangle \theta_{ij}. \end{aligned}$$

Defining

$$Q_{ij}(t) = \exp\{-i(\epsilon_i - \epsilon_j)t\} \langle \theta_{ij}(t) \rangle, \quad (\text{B.6})$$

we have

$$Q_{ij}(t) = \left\langle \exp\left[i \int_0^t V^x(\tau) d\tau\right] \right\rangle \theta_{ij}, \quad (\text{B.7})$$

$$\frac{d}{dt} Q_{ij}(t) = \left\langle \exp\left[i \int_0^t V^x(\tau) d\tau\right] \right\rangle \sum_{n=2}^{\infty} \frac{1}{n!} K_n^x(t) \theta_{ij}. \quad (\text{B.8})$$

To lowest nonvanishing order this becomes,

$$\frac{d}{dt} Q_{ij}(t) = - \left\langle \exp\left[i \int_0^t V^x(\tau) d\tau\right] \right\rangle \int_0^t \langle [V(\tau), [V(t), \theta_{ij}]] \rangle d\tau. \quad (\text{B.9})$$

Using

$$V(t) = \sum_{kl} B_k(t) \mathcal{S}_{kl} \exp(i\omega_l t)$$

in the commutator and using the fact that bath and system operators commute, we can bring (B.9) into the form

$$\frac{d}{dt} Q_{ij}(t) = - \left\langle \exp \left[i \int_0^t V^x(\tau) d\tau \right] \right\rangle \sum_q c_q(t) \mathcal{L}_q \theta_{ij} \mathcal{R}_q, \quad (\text{B.10})$$

where \mathcal{L}_q and \mathcal{R}_q are system operators and $c_q(t)$ is a number obtained by evaluating a bath correlation function

$$c_q(t) = \int_0^t \langle \mathcal{B}_\alpha(\tau) \mathcal{B}_\beta(t) \rangle \exp(i\omega_\alpha \tau) \exp(i\omega_\beta t) d\tau. \quad (\text{B.11})$$

We also could have formally used the density matrix in

$$\int_0^t \langle [V(\tau), [V(t), \rho]] \rangle d\tau$$

in place of θ_{ij} , which would then give us the expression

$$E = \sum_q c_q(t) \mathcal{L}_q \rho \mathcal{R}_q. \quad (\text{B.12})$$

We now continue with the process of finding the equation of motion for θ_{ij} . Consider the operator $\mathcal{L}_q \theta_{ij} \mathcal{R}_q$. Its (kl) matrix element is $(\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl}$. Therefore

$$\mathcal{L}_q \theta_{ij} \mathcal{R}_q = \sum_{kl} (\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl} \theta_{kl}. \quad (\text{B.13})$$

In (B.13), we should be aware that while θ_{kl} is an operator, the multiplying factor $(\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl}$ is simply a number. Therefore using (B.7) and (B.10), we find

$$\frac{d}{dt} Q_{ij}(t) = \sum_q c_q(t) \sum_{kl} (\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl} Q_{kl}(t), \quad (\text{B.14})$$

$$\begin{aligned} \frac{d}{dt} \langle \theta_{ij} \rangle &= i(\varepsilon_i - \varepsilon_j) \langle \theta_{ij} \rangle \\ &+ \sum_q c_q(t) \sum_{kl} (\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl} Q_{kl}(t) \exp \{ i(\varepsilon_i - \varepsilon_j) t \}. \end{aligned} \quad (\text{B.15})$$

Now, we generally have the situation that terms for which $\varepsilon_k - \varepsilon_l \neq \varepsilon_i - \varepsilon_j$ make negligible contributions to the right side of (B.14) because $c_q(t)$ is a strongly oscillating function. Therefore, we can replace $\exp \{ i(\varepsilon_i - \varepsilon_j) t \}$ by $\exp \{ i(\varepsilon_k - \varepsilon_l) t \}$ and write

$$\frac{d}{dt} \langle \theta_{ij} \rangle = i(\varepsilon_i - \varepsilon_j) \langle \theta_{ij} \rangle + \sum_q c_q(t) \sum_{kl} (\mathcal{L}_q)_{ki} (\mathcal{R}_q)_{jl} \langle \theta_{kl} \rangle. \quad (\text{B.16})$$

We now define a tetradic operator T whose matrix elements are

$$T_{ji, ik}^{(a)} = (\mathcal{L}_a)_{kl} (\mathcal{R}_a)_{jl}. \quad (\text{B.17})$$

In terms of these matrix elements, we have

$$\frac{d}{dt} \langle \theta_{ij} \rangle = i(\varepsilon_i - \varepsilon_j) \langle \theta_{ij} \rangle + \sum_a c_a(t) \sum_{kl} T_{ji, ik}^{(a)} \langle \theta_{kl} \rangle. \quad (\text{B.18})$$

Now if $\varrho(0)$ is the density matrix at $t = 0$, we take the trace of the product of ϱ with both sides of this equation

$$\frac{d}{dt} \text{Tr} \varrho \langle \theta_{ij}(t) \rangle = i(\varepsilon_i - \varepsilon_j) \text{Tr} \varrho \langle \theta_{ij}(t) \rangle + \sum_a c_a(t) \sum_{kl} T_{ji, ik}^{(a)} \text{Tr} \varrho \langle \theta_{kl}(t) \rangle. \quad (\text{B.19})$$

However,

$$\text{Tr} \varrho \langle \theta_{ij}(t) \rangle = \text{Tr} \varrho(t) \theta_{ij} = \varrho_{ji}(t).$$

Hence we have the following coupled equations for the matrix elements of $\varrho(t)$

$$\frac{d\varrho_{ji}}{dt} = i(\varepsilon_i - \varepsilon_j) \varrho_{ji}(t) + \sum_a c_a(t) \sum_{kl} T_{ji, ik}^{(a)} \varrho_{lk}(t). \quad (\text{B.20})$$

Since H_S is diagonal in the chosen basis, with $H_{ii} = \varepsilon_i$, we have

$$(\varepsilon_i - \varepsilon_j) \varrho_{ji}(t) = [\varrho(t) H - H \varrho(t)]_{ji}. \quad (\text{B.21})$$

Furthermore we can show that when the tetradic operator defined by (B.17) operates on any ordinary dyadic operator A , the result is

$$\mathcal{T}^{(a)} A = \mathcal{R}_a A \mathcal{L}_a. \quad (\text{B.22})$$

Using (B.21) and (B.22) in (B.20), we find the matrix equation

$$\frac{d\varrho}{dt} = i[\varrho(t), H] + \sum_a c_a(t) \mathcal{R}_a \varrho(t) \mathcal{L}_a, \quad (\text{B.23})$$

which is the form taken by the master equation as derived by the cumulant method. It should be noted that operators other than ϱ appearing in the equation are the time independent operators.

If we compare the irreversible term (second term) of (B.23) with the expression (B.12) which results from substituting ϱ directly into

$$\int_0^t \langle [V(\tau), [V(t), \varrho]] \rangle d\tau, \quad (\text{B.24})$$

we see that in order to go from (B.12) to the irreversible term of the master equation, operators which appear on the left and right of ϱ must be reversed. The prescription for finding the irreversible term then is to evaluate the expression (B.24) and then reverse the positions of operators to the left and right of ϱ .

The irreversible term we obtain may be compared with the corresponding irreversible term in the Zwanzig formalism in the following way. Writing $V(t) = \sum_k S_k(t) B_k(t)$, the irreversible term E may be written¹⁹⁾

$$E_z = -\int_0^t \text{Tr}_R ([V(t), [V(\tau), \varrho(t) f_0(R)]] d\tau, \quad (\text{B.25})$$

where $f_0(R)$ is the reduced density matrix for the bath at $t = 0$. On the other hand, by the cumulant method, we have,

$$E_c = \mathcal{R} E'_c \quad (\text{B.26})$$

where

$$E'_c = -\int_0^t \text{Tr}_R [V(\tau), [V(t), \varrho(t)]] f_0(R) d\tau$$

and \mathcal{R} denotes the operation of reversing operators on either side of ϱ . Substituting the expanded $V(t)$ in both these expressions,

$$\begin{aligned} E_z = & -\sum_{ij} \int_0^t \text{Tr}_R \{ S_i(t) S_j(\tau) \varrho(t) B_i(t) B_j(\tau) f_0(R) \\ & - S_i(t) \varrho(t) S_j(\tau) B_j(\tau) B_i(t) f_0(R) \\ & - S_j(\tau) \varrho(t) S_i(t) B_i(t) B_j(\tau) f_0(R) \\ & + \varrho(t) S_j(\tau) S_i(t) B_j(\tau) B_i(t) f_0(R) \} d\tau, \end{aligned} \quad (\text{B.27a})$$

$$\begin{aligned} E'_c = & -\sum_{ij} \int_0^t \text{Tr}_R \{ S_i(\tau) S_j(t) \varrho(t) B_i(\tau) B_j(t) f_0(R) \\ & - S_i(\tau) \varrho(t) S_j(t) B_i(\tau) B_j(t) f_0(R) \\ & - S_j(t) \varrho(t) S_i(\tau) B_j(t) B_i(\tau) f_0(R) \\ & + \varrho(t) S_j(t) S_i(\tau) B_j(t) B_i(\tau) f_0(R) \} d\tau, \end{aligned} \quad (\text{B.27b})$$

$$\begin{aligned} E_c = & -\sum_{ij} \int_0^t \text{Tr}_R \{ \varrho(t) S_i(\tau) S_j(t) B_i(\tau) B_j(t) f_0(R) \\ & - S_j(t) \varrho(t) S_i(\tau) B_i(\tau) B_j(t) f_0(R) \\ & - S_i(\tau) \varrho(t) S_j(t) B_j(t) B_i(\tau) f_0(R) \\ & + S_j(t) S_i(\tau) \varrho(t) B_j(t) B_i(\tau) f_0(R) \} d\tau. \end{aligned} \quad (\text{B.27c})$$

In writing (B.27a), we have used the cyclic property of the trace to rearrange bath operators. On reversing the dummy indices and the first and last terms in (B.27c), we see that the expression E_c is identically equal to E_z .

Appendix C

This appendix is divided into three sections. In the first, we define a unitary transformation connecting Dicke and locally excited states and use the symmetry properties of spatially small systems in order to evaluate bilinear sums of transformation elements. In this way we arrive at a reduced transformation in which the degeneracy parameter α is summed out. In the second part, we illustrate the application of the reduced transformation to $\langle R^z \rangle$ and other collective operators. Finally, in the last section, the transformation is applied to the problem of obtaining a propagator for the density matrix in terms of locally excited states.

(1) We denote the unitary transformation from the basis of Dicke states to local excitation states by V . We will see in sections (2) and (3) that in many examples of physical interest, we are confronted with certain combinations of the matrix elements of V rather than the individual matrix elements. In the matrix element V_{ps} , the right-hand index refers to a Dicke state; the left, to a local state. Therefore s is really an abbreviation for the usual quantum numbers (r, m, α) used to designate Dicke states. Thus we have functions $r(s)$, $m(s)$, and $\alpha(s)$ such that as s runs through its range, (r, m, α) with $r = r(s)$, $m = m(s)$, and $\alpha = \alpha(s)$ run through the range of a complete set of Dicke states. Therefore it is appropriate to write $s = \{r(s), m(s), \alpha(s)\}$. The sums we evaluate here are

$$E = \sum_{\alpha} V_{l, (r m \alpha)} V_{(r m' \alpha), l'}^+$$

The labels l and l' refer to two states of local excitation. Since these states are associated with definite values of the total energy, we have $F = 0$ unless the energies of $|l\rangle$ and $|l'\rangle$ are m and m' respectively. If this condition is satisfied, we further expect from symmetry considerations that E depends only on m, m', r , and the relatedness $Q(l, l')$. Therefore, reversing indices and performing a complex conjugate operation to get rid of the adjoint in the second factor, we write

$$P_{Q(r, m, m')} = \sum_{\alpha} V_{l, (r m \alpha)} V_{l', (r m' \alpha)}^* \quad (\text{C.1})$$

The first case for which we evaluate this expression is for $l = l'$ ($Q = 0$). We do this by evaluating

$$\Omega_{ll} = \sum_{r m \alpha} \sum_{r' m' \alpha'} V_{l, (r m \alpha)} \theta_{(r m \alpha), (r' m' \alpha')} V_{l, (r' m' \alpha')}^* \quad (\text{C.2})$$

for the operator

$$\theta_{(r m \alpha), (r' m' \alpha')} = \delta_{rr'} \delta_{rrp} \delta_{mm'} \delta_{mm_p} \delta_{\alpha\alpha'}, \quad (\text{C.3})$$

where r_p and m_p are chosen values of the quantum numbers r and m . For this operator,

$$\Omega_{ll} = \sum_{\alpha} V_{l, (r_p m_p \alpha)} V_{l, (r_p m_p \alpha)}^* \quad (\text{C.4})$$

This expression is zero for localized states l with total energy not equal to m_p . However, according to (C.2), when the energy $m(l)$ is equal to m_p , Ω_{ll} is equal to $P_0(r_p, m_p, m_p)$. Since there are $\binom{N}{\frac{1}{2}N - m_p}$ such values of l , the trace of Ω evaluated in the local state basis is just

$$P_0(r_p, m_p, m_p) \binom{N}{\frac{1}{2}N - m_p}. \quad (\text{C.5})$$

On the other hand, the non-vanishing diagonal matrix elements of θ are all equal to 1 and there are $g(r_p)$ of these where $g(r_p)$ is the degeneracy of the state $|r_p, m_p\rangle$. Hence the trace of θ is $g(r_p)$. Therefore by the invariance of the trace, we have

$$P_0(r_p, m_p, m_p) = g(r_p) \left/ \binom{N}{\frac{1}{2}N - m_p} \right. \quad (\text{C.6})$$

In order to evaluate $P_Q(r, m, m')$ for other choices of Q and m' , we develop recurrence relations by expressing the transformation V in terms of the eigenvectors of R^2 and R^z , using the relationship

$$R^- |r, m, \alpha\rangle = (r(r+1) - m(m-1))^{\frac{1}{2}} |r, m-1, \alpha\rangle \quad (\text{C.7})$$

for the normalized eigenvectors, and taking into account the symmetry properties of small systems. If the expansion

$$\sum_l c_l(r, m, \alpha) |l\rangle = |r, m, \alpha\rangle \quad (\text{C.8})$$

is an expression for the normalized Dicke state in terms of localized states, then the general theory of unitary transformation provides that

$$c_l(r, m, \alpha) = V_{(rm\alpha), l}^+ \quad (\text{C.9})$$

As a consequence, we may write

$$P_Q(r, m, m') = \sum_{\alpha} c_l^*(r, m, \alpha) c_l(r, m', \alpha). \quad (\text{C.10})$$

Substitution of (C.8) into (C.7) yields the following relationship between expansion coefficients for different states

$$c_i(r, m-1, \alpha) = \sum_{\substack{i=1 \\ s_i(l) < 0}}^N c_{l(+i)}(r, m, \alpha) (r(r+1) - m(m-1))^{-\frac{1}{2}}, \quad (\text{C.11})$$

where $s_i(l) = \langle l | \sigma_i^z | l \rangle$ and $|l(+i)\rangle$ is the state $\sigma_i^+ |l\rangle$. When (C.11) is substituted for one or both factors on the right-hand side of (C.10), the result is a sum of terms involving expressions like (C.10) but with other values of the parameters Q , m , and m' . Now the original states $|l\rangle$ and $|l'\rangle$ differ in a certain way in the total amount of excitation each contains and in the way this excitation is distributed over the N atoms. Knowledge of the excitation and its distribution enables us to classify the atoms in sets as follows:

- S_e = atoms which are excited in both $|l\rangle$ and $|l'\rangle$,
- S_u = atoms which are unexcited in both $|l\rangle$ and $|l'\rangle$,
- S_q = atoms which are excited in $|l\rangle$ but unexcited in $|l'\rangle$,
- $S_{q'}$ = atoms which are excited in $|l'\rangle$ but unexcited in $|l\rangle$.

The number of atoms in each class determines the value of the parameters m , m' , q , and q' . When we substitute (C.11) for the first factor in (C.10), we must consider the above classification scheme for each of the following pairs of states:

$$|l(+i)\rangle \quad \text{and} \quad |l'\rangle \quad \text{for } i \text{ such that } S_i(l) < 0.$$

Those atoms i satisfying the condition $S_i(l) < 0$ fall into either $S_u(l, l')$ or into $S_{q'}(l, l')$. For those atoms i in $S_u(l, l')$, we see that the i th atom classified with respect to the new pair of states ($|l(+i)\rangle$, $|l'\rangle$) now falls in the set $S_q(l(+i), l')$ while all other atoms retain their classifications. Similarly if the i th atom is in $S_{q'}(l, l')$, its new classification is $S_e(l(+i), l')$. Now $P_Q(r, m, m')$ is only a function of r and of the numbers n_e , n_u , n_q , and $n_{q'}$ which represent the respective sizes of the sets S_e , S_u , S_q , and $S_{q'}$,

$$f(r, n_e, n_u, n_q, n_{q'}).$$

Substituting (C.11) for the first factor in (C.10), we have

$$P_Q(r, m, m') = \sum_{\substack{i=1 \\ s_i(l) < 0}}^N \sum_{\alpha} \frac{c_{l(+i)}^*(r, m+1, \alpha) c_{l'}(r, m', \alpha)}{(r(r+1) - m(m+1))^{\frac{1}{2}}}. \quad (\text{C.12})$$

Then the above argument about the reclassification of the i th atom leads to the recurrence relation

$$\begin{aligned}
 f(r, n_e, n_u, n_q, n_{q'}) &= \frac{1}{(r(r+1) - m(m+1))^{\frac{1}{2}}} \\
 &\times \{n_u f(r, n_e, n_u - 1, n_q + 1, n_{q'}) \\
 &+ n_{q'} f(r, n_e + 1, n_u, n_q, n_{q'} - 1)\}. \quad (C.13)
 \end{aligned}$$

Since $n_e + n_q = \frac{1}{2}N + m$, $n_e + n_{q'} = \frac{1}{2}N + m'$, and $n_q + n_{q'} = Q$, we may express the recurrence relation in terms of $P_Q(r, m, m')$ as

$$\begin{aligned}
 P_Q(r, m, m') &= \frac{1}{(r(r+1) + m(m+1))^{\frac{1}{2}}} \\
 &\times \left\{ \frac{1}{2}(Q + m' - m) P_{Q-1}(r, m + 1, m') \right. \\
 &\left. + \frac{1}{2}(N - Q - m - m') P_{Q+1}(r, m + 1, m') \right\}. \quad (C.14)
 \end{aligned}$$

We have derived this relationship first because it was more easily discussed. However, in order to establish properly contact with the result (C.6) we also need recurrence relations which arise from considering (C.10) with $m = m'$ and substituting (C.11) for *both* factors on the right side. The argument is similar to the one leading to (C.14) and leads to the recurrence formula

$$\begin{aligned}
 P_{q+1}(r, m + 1, m + 1) &= \frac{1}{a^2 - a} \{ [r(r+1) - m(m+1)] P_q(r, m, m) \\
 &- a(2q + 1) P_q(r, m + 1, m + 1) \\
 &- q^2 P_{q-1}(r, m + 1, m + 1) \} \quad (C.15)
 \end{aligned}$$

where $a = \frac{1}{2}N - m - q$, q as defined in text: $q = q' = \frac{1}{2}Q$. Using the expression (C.6) with $q = 0$ to start the recurrence, we solve the relations (C.15) for $P_Q(r, m, m)$. Then we use the resulting form to start the recurrence relation (C.14) and obtain $P_Q(r, m', m)$. The result is

$$\begin{aligned}
 P_Q(r, m', m) &= \left(\frac{(r + m')!(r + m)!}{(r - m')!(r - m)!} \right)^{\frac{1}{2}} \frac{g(r)}{N!} (\frac{1}{2}N - m - q)! q'! \\
 &\times \sum_{j=0}^r (-1)^j \binom{q}{j} \frac{(r - m + q' - j)!}{(q' - j)!} \frac{(\frac{1}{2}N + m + j - q')!}{(r + m + j - q')!}. \quad (C.16)
 \end{aligned}$$

When $r = \frac{1}{2}N$, the expression (C.1) has only one term, which is equal to

$$\frac{1}{N!} [(\frac{1}{2}N + m)! (\frac{1}{2}N - m')! (\frac{1}{2}N - m)! (\frac{1}{2}N + m')!]^{\frac{1}{2}}. \quad (\text{C.17})$$

In this case, the last factor in each term of (C.16) is unity and the sum may be evaluated in closed form since it represents the q th order difference of a polynomial of the form $x(x+1)\cdots(x+r-m-1)$. The result agrees with (C.17) for all values of q . Another case permitting easy evaluation of the summation is $r = m$. The result is

$$P_Q(m, m', m) = \binom{2m}{m+m'}^{\frac{1}{2}} \binom{\frac{1}{2}N+m}{\frac{1}{2}N+m'-q}^{-2} \frac{2m+1}{(\frac{1}{2}N+m+1)}. \quad (\text{C.18})$$

(2) We illustrate here the use of the reduced transformation in the evaluation of certain collective operators. Consider first the operator R^2 . Here

$$\theta_{(rm\alpha), (r'm'\alpha')} = r(r+1) \delta_{rr'} \delta_{mm'} \delta_{\alpha\alpha'}.$$

Therefore, the matrix corresponding to R^2 in the basis of local states is, according to (C.2)

$$\begin{aligned} \Omega_{ll'} &= \sum_{r, \alpha} V_{l, (rm\alpha)} r(r+1) V_{l', (rm\alpha)}^* \\ &= \sum_{r=|m|}^{\frac{1}{2}N} r(r+1) P_{Q(l, l')}(r, m, m). \end{aligned} \quad (\text{C.19})$$

Here $\Omega_{ll'}$ vanishes for states of differing m value. The value m on the right side is the common m value associated with l and l' . In particular we find for $l = l'$ that

$$\Omega_{ll} = \binom{N}{\frac{1}{2}N - m}^{-1} \sum_{r=|m|}^{\frac{1}{2}N} r(r+1) g(r). \quad (\text{C.20})$$

As a second example, consider the thermally averaged operators $\langle\langle (R^2)^n(t) \rangle\rangle$. The form of the coupled moment equations (2.15) implies that since R^2 and $\langle\langle (R^2)^n \rangle\rangle$ are initially diagonal in the basis of Dicke states, they always remain diagonal. In this case, we have to consider operators of the form

$$\theta_{(rm\alpha), (r'm'\alpha')} = f(r, m, t) \delta_{rr'} \delta_{mm'} \delta_{\alpha\alpha'},$$

where, for example, for θ corresponding to $\langle\langle (R^2)^n \rangle\rangle$, $f(r, m, t)$ may be found either by solving the coupled moment equations (2.15) subject to the initial condition

$$\{R^2, \langle R^2 \rangle, \langle\langle (R^2)^2 \rangle\rangle, \dots\} = \{r(r+1), m, m^2, \dots\}$$

for $\langle (R^z)^n \rangle$ or by taking the appropriate moment of the master eq. (3.5) with $m = m', r = r', \alpha = \alpha'$, subject to $Q_{rm\alpha, rm\alpha} = 1$. In this case

$$\Omega_{ll'}(t) = \sum_{r=|m|}^{\frac{1}{2}N} f(r, m, t) P_{Q(l, l')}(r, m, m). \quad (\text{C.21})$$

In particular, for $l = l'$, $\Omega_{ll}(t)$ is the expectation value of $\langle R^z \rangle$ in a system initially in a locally excited state $|l\rangle$.

(3) We now consider the application of the transformation to the propagator which contains information about the energy transfer from one part of the system to another. Just as the propagator for the wavefunction is a dyadic operator, the propagator for the density matrix is a tetradic. Dyadic operators are transformed according to

$$\Omega = V\theta V^+ \quad (\text{C.22})$$

or

$$\Omega = \mathcal{V}\theta, \quad (\text{C.23})$$

where \mathcal{V} is a tetradic operator related to V by

$$\mathcal{V}_{ll', jk} = V_{lj} V_{kl'}^+ = V_{lj} V_{l'k}^*. \quad (\text{C.24})$$

Tetradic operators, then, such as T defined by (3.10), transform into the basis of local states according to

$$\mathcal{T} = \mathcal{V}T\mathcal{V}^+. \quad (\text{C.25})$$

This follows since the unitarity of V implies that \mathcal{V} is also unitary. Incorporating (C.24) into (C.25), we find the explicit matrix element expression

$$\begin{aligned} \mathcal{T}_{ij, kl} = & \sum_{u, v, w, x} V_{iu} \delta_{m_u m_l} V_{jv}^* \delta_{m_v m_j} T_{uv, wx}(r_u, r_v, m, t) \\ & \times \delta_{r_u r_w} \delta_{r_v r_x} \delta_{\alpha_u \alpha_w} \delta_{\alpha_v \alpha_x} V_{kw}^* \delta_{m_w m_k} V_{lx} \delta_{m_x m_l}. \end{aligned} \quad (\text{C.26})$$

Here $\mathbf{m} = (m_u, m_v, m_w, m_x)$. The dummy indices all refer to Dicke states $u = (r_u, m_u, \alpha_u)$, $v = (r_v, m_v, \alpha_v)$, etc. Taking into account the δ functions,

$$\begin{aligned} \mathcal{T}_{ij, kl} = & \sum_{r_u=M_u}^{\frac{1}{2}N} \sum_{r_v=M_v}^{\frac{1}{2}N} \sum_{\alpha_u=1}^{g(r_u)} \sum_{\alpha_v=1}^{g(r_v)} V_{l(u)}(r_u, \alpha_u, m_l) \\ & \times V_{j(v)}(r_v, \alpha_v, m_j) T(r_u m_l, r_v m_j; r_u m_k, r_v m_l) \\ & \times V_{k(w)}^*(r_u, \alpha_u, m_k) V_{l(k)}(r_v, \alpha_v, m_l), \end{aligned} \quad (\text{C.27})$$

where

$$M_u = \max \{ \text{abs } m_i, \text{abs } m_k \},$$

$$M_v = \max \{ \text{abs } m_j, \text{abs } m_l \}.$$

Since T does not depend on α_u or α_v , this becomes

$$\begin{aligned} \mathcal{F}_{ij,kl} &= \sum_{r_u=M_u}^{\frac{1}{2}N} \sum_{r_v=M_v}^{\frac{1}{2}N} T(r_u m_i, r_v m_j; r_u m_k, r_v m_l) \\ &\quad \times \sum_{\alpha_u=1}^{g(r_u)} V_i(r_u, \alpha_u, m_i) V_k(r_u, \alpha_u, m_k) \\ &\quad \times \sum_{\alpha_v=1}^{g(r_v)} V_j^*(r_v, \alpha_v, m_j) V_l(r_v, \alpha_v, m_l) \\ &= \sum_{r_u=M_u}^{\frac{1}{2}N} \sum_{r_v=M_v}^{\frac{1}{2}N} T(r_u m_i, r_v m_j; r_u m_k, r_v m_l) \\ &\quad \times P_{Q(i,k)}(r_u, m_i, m_k) P_{Q(j,l)}(r_v, m_j, m_l). \end{aligned} \tag{C.28}$$

Therefore we can transform the propagator for ϱ using the expressions $P_Q(r, m', m)$ developed earlier in (C.16).

References

- 1) R.H. Dicke, Phys. Rev. **93** (1954) 99.
- 2) G.S. Agarwal, Springer Tracts in Modern Physics **70** (1974) 1 and references contained therein.
- 3) F. Haake, Springer Tracts in Modern Physics **66** (1973) and references contained therein.
- 4) R. Friedberg and S.R. Hartmann, Phys. Rev. **A10** (1974) 1728.
- 5) F. Haake and R.J. Glauber, Phys. Rev. **A5** (1972) 1457.
G. Oliver and A. Tallet, Phys. Rev. **A7** (1973) 1061.
- 6) R. Bonifacio, P. Schwendimann and F. Haake, Phys. Rev. **A4** (1971) 302, 854.
R. Bonifacio and A. Lugiato, Phys. Rev. **A11** (1975) 1507.
- 7) P.S. Lee, Y.C. Lee and C.T. Chang, Phys. Rev. **A8** (1973) 1722.
P.S. Lee and Y.C. Lee, Phys. Rev. **A8** (1973) 1727.
- 8) N.E. Rehler and J.H. Eberly, Phys. Rev. **A3** (1971) 1735.
- 9) V. Degiorgio and F. Ghielmetti, Phys. Rev. **A4** (1971) 2415.
- 10) R. Bonifacio and P. Schwendimann, Lett. Nuovo Cimento **3** (1970) 509, 512.
- 11) W.H. Louisell, Quantum Statistical Properties of Radiation (John Wiley, New York, 1973).
- 12) G.I. Peters and L. Allen, J. Phys. **A4** (1974) 238.
- 13) R.H. Picard and C.R. Willis, Phys. Rev. **A8** (1973) 1536.

- 14) A. Nitzan and R. J. Silbey, *J. Chem. Phys.* **60** (1974) 4070; note that eq. (10) in this reference is in error since the time-ordering in the exponential is antichronological (*i.e.*, $t_{\text{left}} < t_{\text{right}}$). This reversed ordering affects the sign of frequency shifts.
- 15) R. Kubo, *J. Phys. Soc. of Japan* **17** (1962) 1100.
- 16) See also B. Yoon, J. M. Deutch and J. H. Freed, *J. Chem. Phys.* **62** (1975) 4687 for a general discussion of the relationship between cumulant and projector methods.
- 17) G. S. Agarwal, *Phys. Rev.* **A2** (1971) 2038.
- 18) G. S. Agarwal, *Phys. Rev.* **A4** (1971) 1791.
- 19) H. J. Carmichael and D. F. Walls, *J. Phys.* **A6** (1973) 1552.