Master-Equation Approach to Spontaneous Emission. III. Many-Body Aspects of Emission from Two-Level Atoms and the Effect of Inhomogeneous Broadening*

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A general theory of spontaneous emission was developed in Papers I and II of this series. In the present paper, the master equation describing spontaneous emission from N identical two-level atoms is investigated further. It is shown that the equation of motion for the $p(<N)$-atom density matrix is coupled to the $(p+1)$-atom density matrix. We first consider the initial excitation of the system to state $|\theta_0, \varphi_0\rangle$ $(\theta_0<\pi)$ and solve the hierarchy of equations so obtained in the "uncorrelated approximation." It is found that the presence of other atoms is equivalent to an external field which is determined from a self-consistent analysis. It is also found that in this approximation the state of the atom at time t can be described by a single parameter $\theta(t)$ $(\theta<\pi)$, with $\sin^2\frac{1}{2}\theta$ giving the probability that the atom is to be found in the excited state. Next, some improvements over the uncorrelated approximation are given. This is done by decoupling the equation of motion for higher-order mean values. It is found that the fluctuation $\langle(S_i^z-\langle S_i^z\rangle)(S_j^z-\langle S_j^z\rangle)\rangle$ for any pair of atoms is of the order of $1/N$. The case when $\theta_0=\pi$ is considered separately and we obtain an expression for the radiation rate by making a "Hartree-Fock" type of approximation on the two-particle mean values. In this case, the behavior of the radiation rate is found to differ markedly from the "sech" behavior. It is then shown that the entire dynamics of two-level atoms emitting spontaneously can be described by a set of $2N$ coupled first-order equations which clearly exhibit the type of nonlinearity (which is the analog of the van der Pol type of nonlinearity) for this problem and provide a better understanding of spontaneous emission. Finally in Sec. V, the theory is extended to include the effects of inhomogeneous broadening, and the functional dependence of the radiation rate on the atomic-line-shape factor is obtained.

I. INTRODUCTION

In Papers I and II of this series,[1,2] we developed a general theory of spontaneous emission from two systems, namely, from a system of N identical two-level atoms and from a system of N identical harmonic oscillators. This general theory employed the standard methods of nonequilibrium statistical mechanics.[3] It was found that the reduced density operator, for the atomic system (whose size is small compared to a wavelength), satisfies the following master equation [Eq. (I A7)]:

$$\frac{\partial\rho}{\partial t}=-\gamma_0\sum_{i,j}(S_i^+S_j^-\rho-2S_j^-\rho S_i^++\rho S_i^+S_j^-)\,, \qquad (1.1)$$

where $2\gamma_0$ is the inverse of the spontaneous lifetime of a single atom and $S_i^\pm$ and S_i^z are the components of the spin angular momentum operators for the ith atom.[4] Equation (1.1), in terms of the collective operators $S^\pm$, S^z introduced by Dicke,[5] can be written as[6]

$$\frac{\partial\rho}{\partial t}=-\gamma_0(S^+S^-\rho-2S^-\rho S^++\rho S^+S^-), \qquad (1.2)$$

where

$$S^\pm=\textstyle\sum_i S_i^\pm,\quad S^z=\sum_i S_i^z\,. \qquad (1.3)$$

In I, we presented the *exact* solution of the master equation (1.2) [cf. Eq. (I 4.7)]. However in any practical solution (for which N is large, say of the order of 10^6) the exact solution is too involved to be of any use. One should therefore resort to approximate methods. It is not clear how the approximations can be made on the master equation involving the collective operators. Moreover we would like to study how the individual atoms behave in spontaneous emission. For instance, one can ask the following question: Are there any correlations induced between any pair of atoms owing to spontaneous emission? We already provided an answer to such a question in II for the case in which each of the two-level atoms is replaced by a harmonic oscillator. For the harmonic-oscillator model, we showed that *no* correlation is induced between any two oscillators if the system was initially excited to a coherent state [superradiant state for this system; cf. Eqs. (II 3.21) and (II 3.22)]. For these reasons it seems better to work with the master equation (1.1) which gives us the many-body description of spontaneous emission and solve it under various approximate schemes.

In Sec. II of the present paper, we use the master equation (1.1) to obtain the equation of motion for the $p(<N)$-atom density matrix.[7] The equation for the p-atom density matrix is found to be coupled to that of $(p+1)$-atom density matrix. We consider the case in which the *atomic system was prepared*

in a state $|\theta_0, \varphi_0\rangle$ [cf. Eq. (2.6)] and make the uncorrelated approximation, i.e., we assume that the total density operator factorizes in terms of the density operators of individual atoms. We then find that the state of each atom at time t can be described by one parameter $\theta(t)$, where the probability amplitude that the atom be in the excited state is $\sin\frac{1}{2}\theta e^{-i\varphi/2}$. The phase φ is found to be a constant of motion. In Sec. III, we obtain a result[8] for the radiation rate that is an improvement over the one obtained by making the uncorrelated approximation. This is done by decoupling the equation of motion for the higher-order mean values. It is shown that, for the case of initial excitations given by $|\theta_0, \varphi_0\rangle$, $(\theta_0 < \pi)$, the fluctuations $\langle S_i^z S_j^z\rangle - \langle S_i^z\rangle\langle S_j^z\rangle$ is of the order $1/N$. In this section we also consider the case in which each of the atoms was *initially in its excited state* and obtain an expression for the radiation rate which differs markedly from the "sech" behavior. In Sec. IV we present a new description of spontaneous emission which is similar to the one used for the description of lasers.[9] This description provides us with a new insight into the process of spontaneous emission. It is found that a complete solution to the problem of spontaneous emission is equivalent to the solution of $2N$ coupled van der Pol-type equations.[10] These equations clearly exhibit the type of nonlinearity which exists for the problem of spontaneous emission from two-level atoms. Finally, in Sec. V we consider the effects of inhomogeneous broadening and derive the appropriate master equation. We then obtain the explicit functional dependence of the radiation rate on the lineshape factor for the atomic system.

II. HIERARCHY OF EQUATIONS FOR P-PARTICLE DENSITY MATRIX AND SOLUTION IN UNCORRELATED APPROXIMATION

In this section, we first consider the many-body problem as described by the master equation (1.1) and obtain the hierarchy of equations for the p-particle density matrix.[7] The p-particle $(p < N)$ density matrix, denoted by ρ_p, is obtained from the N-particle density matrix by taking the trace over the coordinates of rest of the particles, i.e.,

$$\rho_p = \mathrm{Tr}_{p+1,p+2,\ldots,N}(\rho)\,, \tag{2.1}$$

where $\mathrm{Tr}_{p+1,p+2,\ldots,N}$ indicates the trace over the coordinates of the $(p+1)$, $(p+2)$, $\ldots$, Nth atoms. On combining (1.1) and (2.1), we obtain the following equation:

$$-\frac{1}{\gamma_0}\frac{\partial\rho_p}{\partial t} = \sum_{1\le i,j\le N} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+ + \rho S_i^+ S_j^-\}\,. \tag{2.2}$$

This equation may be rewritten as

$$\begin{aligned}-\frac{1}{\gamma_0}\frac{\partial\rho_p}{\partial t} = {}& \sum_{1\le i,j\le p} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+ + \rho S_i^+ S_j^-\} + \sum_{p+1\le i\le N} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_i^-\rho - 2S_i^-\rho S_i^+ + \rho S_i^+ S_i^-\}\\ &+ \sum_{1\le i\le p,\, p+1\le j\le N} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+ + \rho S_i^+ S_j^- + S_j^+ S_i^-\rho - 2S_i^-\rho S_j^+ + \rho S_j^+ S_i^-\}\\ &+ \sum_{p+1\le i<j\le N} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+ + \rho S_i^+ S_j^- + S_j^+ S_i^-\rho - 2S_i^-\rho S_j^+ + \rho S_j^+ S_i^-\}\,.\end{aligned} \tag{2.3}$$

It follows from the cylic property of the trace that the terms in the second summation and in the last summation on the right-hand side of (2.3) are identically equal to zero. Then (2.3) reduces to

$$\begin{aligned}-\frac{1}{\gamma_0}\frac{\partial\rho_p}{\partial t} = {}& \sum_{1\le i,j\le p}\{S_i^+ S_j^-\rho_p - 2S_j^-\rho_p S_i^+ + \rho_p S_i^+ S_j^-\}\\ &+ \sum_{1\le i\le p,\, p+1\le j\le N} \mathrm{Tr}_{p+1,p+2,\ldots,N}\{S_i^+ S_j^-\rho - 2S_j^-\rho S_i^+ + \rho S_i^+ S_j^- + S_j^+ S_i^-\rho - 2S_i^-\rho S_j^+ + \rho S_j^+ S_i^-\}\,.\end{aligned} \tag{2.4}$$

We now simplify the last term on the right-hand side of (2.4). Since the atoms are identical, it follows that all the terms in $\sum_{p+1\le j\le N}$ will give identical contribution. Then on introducing the $(p+1)$-particle density matrix ρ_{p+1}, defined by a relation similar to (2.1), we find that (2.4) reduces to

$$\begin{aligned}\frac{\partial\rho_p}{\partial t} = {}& -\gamma_0 \sum_{i,j=1}^{p}(S_i^+ S_j^-\rho_p - 2S_j^-\rho_p S_i^+ + \rho_p S_i^+ S_j^-)\\ &- \gamma_0(N-p)\,\mathrm{Tr}_{p+1}\{S_{p+1}^-[\sum_{i=1}^{p} S_i^+, \rho_{p+1}] + \mathrm{H.c.}\}\,.\end{aligned} \tag{2.5}$$

We thus find that the equation of motion for the p-particle density matrix is coupled to that for the $(p+1)$-particle density matrix, and so on. This is a general feature of the many-particle systems. In order to obtain a closed set of equations, we must

truncate the hierarchy by making a suitable approximation on the higher-order density matrix.

We will assume that the system was initially prepared in a state which is free of "correlations" and for which the dipole moment has a "finite value." In particular we assume that the state (which can be obtained by appropriate excitation by external fields) at time $t=0$ is given by

$$\rho(0)=\prod_i |\theta_0, \varphi_0\rangle_{i\,i}\langle\theta_0, \varphi_0| , \quad \theta_0<\pi , \tag{2.6}$$

where

$$|\theta_0, \varphi_0\rangle_i = \sin\tfrac{1}{2}\theta_0\, e^{-i\varphi_0/2}\,|+\rangle_i + \cos\tfrac{1}{2}\theta_0\, e^{i\varphi_0/2}|-\rangle_i . \tag{2.7}$$

Here θ_0 and φ_0 are two parameters which characterize the initial state, and $|+\rangle_i$ and $|-\rangle_i$ are the excited and the ground states of the ith atom. Then it can be easily shown that the radiation rate, in first-order perturbation theory, is given by [cf. also Eq. (2.18)]

$$I=2\gamma_0\omega_0 N[1+(N-1)\cos^2\tfrac{1}{2}\theta_0]\sin^2\tfrac{1}{2}\theta_0 . \tag{2.8}$$

Equation (2.8) shows that for large values of N, I is proportional to N^2 when $\theta_0=\frac{1}{2}\pi$, i.e., the system when excited to the state (2.6) with $\theta_0=\frac{1}{2}\pi$ leads to the "superradiant emission" even though there are "no correlations" among different atoms. The important point to notice here is that the dipole moment takes maximum value for such a situation. On the other hand, as we will see later, a system in a state with "zero dipole moment" and with "no correlations" *cannot* lead to superradiant emission. We also recall that if each two-level atom is replaced by a harmonic oscillator then, as we saw in II [Eqs. (II 3.21) and (II 3.22)], the total density matrix (when each of the oscillators was excited to a superradiant state) can be expressed as a product of one-particle density matrices. In view of these facts, it appears reasonable to assume that the p-particle density matrix can be written as a product of one-particle density matrices. We will refer to this approximation as the "uncorrelated approximation," i.e.,

$$\rho_p=\prod\rho_1 . \tag{2.9}$$

Then the equation of motion for the one-particle density matrix ρ_1 becomes

$$\frac{\partial\rho_1}{\partial t}=-\gamma_0(S_1^+S_1^-\rho_1-2S_1^-\rho_1S_1^++\rho_1S_1^+S_1^-) - \gamma_0(N-1)\{\langle S_1^-\rangle[S_1^+,\rho_1]+\text{H.c.}\} . \tag{2.10}$$

Here $\langle S_1^\pm\rangle$ are, of course, the one-particle expectation values. It is seen that the equation for the one-particle density matrix, in the uncorrelated approximation, is the same as would be obtained for a damped driven two-level atom. Here the driving field is to be determined from a self-consistent analysis. It should be noted that the one-particle density matrix can also be expressed as

$$\rho_1(t)=\tfrac{1}{2}+2\langle S_1^z(t)\rangle S_1^z+\langle S_1^+(t)\rangle S_1^-+\langle S_1^-(t)\rangle S_1^+ . \tag{2.11}$$

Moreover, since all the atoms are identical and they are initially excited to a permutation symmetric state [Eq. (2.6)], the expectation values $\langle S_1^z(t)\rangle$, $\langle S_1^\pm(t)\rangle$ will be independent of the labeling. On combining (2.10) and (2.11) we obtain

$$\frac{\partial s^*}{\partial t}=-2\gamma_0(s^z+\tfrac{1}{2})-2\gamma_0(N-1)|s|^2 , \qquad \frac{\partial s}{\partial t}=-\gamma_0 s+2\gamma_0(N-1)ss^z , \tag{2.12}$$

where

$$\langle S_1^z\rangle=s^z , \quad \langle S_1^-\rangle=s , \quad \langle S_1^+\rangle=s^* . \tag{2.13}$$

It follows from the second of Eqs. (2.12) that if $s(t=0)=0$, then $s(t)=0$, which implies that the radiation rate is proportional to the number of atoms, which confirms the statement made earlier, viz., the atoms in a state that has zero dipole moment and that has no correlations do not give rise to superradiant emission.

We now consider the superradiant case. We note that the first terms on the right-hand side of Eqs. (2.12) lead to normal emission and these terms appear to be important only for times less than $1/\gamma_0$. Hence in what follows we will consider the solution for times greater than $1/\gamma_0$ and we will ignore the effect of these terms since we are only considering the superradiant emission. It then follows from (2.12) that

$$\frac{\partial}{\partial t}\{(s^z)^2+|s|^2\}=0 \quad \text{or} \quad (s^z)^2+|s|^2=\tfrac{1}{4} , \tag{2.14}$$

where we have used the initial condition (2.6). This result enables us to represent the expectation value in the form

$$\langle S_1^\pm(t)\rangle=\tfrac{1}{2}\sin\theta(t)\,e^{\pm i\varphi(t)}, \quad \langle S_1^z(t)\rangle=-\tfrac{1}{2}\cos\theta(t) . \tag{2.15}$$

On combining (2.12) and (2.15) it can be shown that $\varphi(t)$ is a constant of motion and $\cos\theta(t)$ is given by

$$\cos\theta(t)=1-2[1+\cot^2\tfrac{1}{2}\theta_0\, e^{2\gamma_0(N-1)t}]^{-1} . \tag{2.16}$$

On combining (2.11) and (2.15) we obtain the following result for the one-particle density matrix:

$$\begin{aligned}\rho_1(t)&=\tfrac{1}{2}-\cos\theta(t)\,S_1^z+\{\tfrac{1}{2}\sin\theta(t)\,e^{i\varphi_0}S_1^-+\text{H.c.}\}\\ &=\tfrac{1}{2}|+\rangle_{1\,1}\langle+|[1-\cos\theta(t)]+\tfrac{1}{2}|-\rangle_{1\,1}\langle-|[1+\cos\theta(t)]\\ &\quad+\{\tfrac{1}{2}|-\rangle_{1\,1}\langle+|\sin\theta(t)\,e^{i\varphi_0}+\text{H.c.}\}\\ &=|\varphi(t),\varphi_0\rangle_{1\,1}\langle\theta(t),\varphi_0| ,\end{aligned} \tag{2.17}$$

where $|\theta(t),\varphi_0\rangle$, is given by (2.7) with θ_0 replaced by $\theta(t)$ which is obtained from (2.16). We have

thus shown that, in the uncorrelated approximation and with the effects of normal emission ignored, the system remains in a state of the form (2.6) if it is initially excited to such a state. A similar (exact) result was found in II for the case of emission from a system of harmonic oscillators. It is interesting to note that the solution can be described by one parameter, namely, $\theta(t)$. The approximations that we have considered are in a sense equivalent to the "semiclassical limit" of the master equation (1.1) for one can adopt the following viewpoint[11]: An initial dipole moment radiates a field which causes the dipole moment of the system to change which in turn leads to an enhancement of the radiation rate. We also remark that in a recent publication of Eberly and Rehler,[12] the solution of the form (2.17) for all times was assumed *a priori*. Our deviation, under the approximations that we stated above, provides the first explicit demonstration that such states in fact occur. It is, of course, desirable to examine the validity of the uncorrelated approximation. This question will be examined in Sec. III.

The superradiant contribution to the radiation rate is given by

$$I_S(t) = 2\gamma_0\omega_0 N(N-1)\langle S_i^+ S_j^-\rangle = \tfrac{1}{2}\gamma_0\omega_0 N(N-1)\sin^2\theta(t)$$
$$= 2\gamma_0\omega_0 N(N-1)[1+\cot^2\tfrac{1}{2}\theta_0\, e^{2\gamma_0(N-1)t}]^{-2}$$
$$\times\cot^2\tfrac{1}{2}\theta_0\, e^{2\gamma_0(N-1)t}\ . \quad (2.18)$$

We will compare this result with the corresponding result obtained from the solution of Eq. (I 3.13) (with $\mu = 1 - 1/N$) for the initial excitation (2.6) [cf. Eq. (3.7)]:

$$I_S(t) = 2\gamma_0\omega_0 N(N-1)[1+\cot^2\tfrac{1}{2}\theta_0\, e^{2\gamma_0 N t}+\Delta t]^{-2}$$
$$\times[\cot^2\tfrac{1}{2}\theta_0\, e^{2\gamma_0 N t}+\Delta t],\quad \Delta t = (1/N)(e^{2\gamma_0 N t}-1)\ . \quad (2.19)$$

A comparison of (2.18) and (2.19) shows that they agree up to terms of order $1/N$ provided that we assume that $N\cot^2\frac{1}{2}\theta_0 \gg 1$. (We recall that we are considering only times greater than $1/\gamma_0$.) This is consistent with the fact that our theory is correct to terms of order $1/N$ since as it is shown explicitly in Sec. III, the fluctuation $\langle S_i^Z S_j^Z\rangle - \langle S_i^Z\rangle\langle S_j^Z\rangle$, which is ignored in the present section, is of the order $1/N$.

It has already been discussed in I and II and in other publications[13] that the properties of the radiation field are determined from the averages involving the collective operators S^+ and S^Z defined by (1.3). A calculation similar to that carried out in II (cf. Sec. IV) shows that the positive frequency part of the electric field operator in the radiation zone is given by

$$E^{(+)}(\vec{r},t) \sim (\omega_0^2/c^2)\{\vec{d}-\hat{n}(\vec{d}\cdot\hat{n})\}$$
$$\times(e^{ik_0 r - i\omega_0 t}/r)\, S^-(t-r/c), \quad (2.20)$$

where $\hat{n}$ is the unit vector in the direction of observation and all other symbols have the usual meaning. We will now give a formula which enables us to calculate the expectation values of the form

$$\langle [S^+(t)]^n [S^-(t)]^m\rangle \equiv \Gamma_{n,m}\ . \quad (2.21)$$

We introduce the generating function $M(x, y)$ defined by

$$M(x,y) = \langle e^{xS^+} e^{yS^-}\rangle\ . \quad (2.22)$$

On substituting (1.3) and on using the uncorrelated approximation (2.9), we find that (2.22) reduces to

$$M(x,y) = \prod_i \langle (1 + xS_i^+ + yS_i^- + xyS_i^+S_i^-)\rangle$$
$$= \prod_i \langle (1+\tfrac{1}{2}xy) + xS_i^+ + yS_i^- + xyS_i^Z\rangle$$
$$= \sin^{2N}\tfrac{1}{2}\theta\{1+[xe^{i\varphi_0}+\cot\tfrac{1}{2}\theta]$$
$$\times[ye^{-i\varphi_0}+\cot\tfrac{1}{2}\theta]\}^N\ . \quad (2.23)$$

On substituting $\theta(t)$, as given by (2.16), we easily obtain the time dependence of the generating function. The correlation functions $\Gamma_{n,m}$ are then obtained from the formula

$$\Gamma_{n,m} = \frac{\partial^{n+m}}{\partial x^n \partial y^m} M(x,y)\Big|_{x=y=0}$$
$$= \sin^{2N}\tfrac{1}{2}\theta\, e^{i(n-m)\varphi_0}\frac{\partial^{n+m}}{\partial x^n\partial y^m}(1+xy)^N\Big|_{x=y=\cot 1/2\theta}$$
$$= \sin^{2N}\tfrac{1}{2}\theta\, e^{i(n-m)\varphi_0}\sum_{l=0}^{N}\frac{N!\,l!}{(N-l)!\,(l-n)!\,(l-m)!}$$
$$\times(\cot\tfrac{1}{2}\theta)^{2l-m-n}\ . \quad (2.24)$$

A special case of $\Gamma_{n,m}$ corresponding to $n = m = 2$ has been recently computed.[13(c)]

We conclude this section by briefly outlining the approach which takes into account the correlations between various atoms. We use the Eq. (2.5) for $p = 2$ and make the approximation $\rho_3 = \rho_2\rho_1$, i.e., we assume that the three-particle density matrix can be written as a product of two-particle and one-particle density matrices. We then obtain the following closed set of equations of motion:

$$\frac{\partial\rho_1}{\partial t} = -\gamma_0(S_1^+S_1^-\rho_1 - 2S_1^-\rho_1 S_1^+ + \rho_1 S_1^+S_1^-)$$
$$-\gamma_0(N-1)\,\mathrm{Tr}_2\{S_2^-[S_1^+,\rho_2]+\mathrm{H.c.}\}\ , \quad (2.25)$$

$$\frac{\partial\rho_2}{\partial t} = -\gamma_0\sum_{i,j=1}^{2}(S_i^+S_j^-\rho_2 - 2S_i^-\rho_2 S_j^+ + \rho_2 S_i^+S_j^-)$$
$$-\gamma_0(N-2)\{\langle S_1^-\rangle[S_1^+ + S_2^+,\rho_2]+\mathrm{H.c.}\}\ . \quad (2.26)$$

The solution to these equations will indicate the way in which the correlations between different atoms develop during the process of spontaneous emission, when the system was *initially excited to* the state (2.6). We again point out that the approximation $\rho_3 = \rho_2\rho_1$ will not lead to the superradiant emission if the system was initially excited to a state with zero dipole moment. In this case one must employ some type of nonlinear decoupling and in Sec. III B we will give one example of such a decoupling.

III. RADIATION RATE

In this section, we will determine the radiation rate by making approximations that are much weaker than the ones given in Sec. II. On using (1.1), we easily find that $\langle S_i^z\rangle = \langle S_i^+S_i^-\rangle - \frac{1}{2}$ satisfies the following equation:

$$\frac{\partial}{\partial t}\langle S_i^z\rangle + \gamma_0\sum_j(\langle S_i^+S_j^-\rangle + \langle S_j^+S_i^-\rangle) = 0 \ . \tag{3.1}$$

By making use of the permutation symmetry it has been shown in I that $\langle \vec{S}_i, \vec{S}_j\rangle = \frac{1}{4}$. On using this result in (3.1), we obtain the equation

$$\frac{\partial}{\partial t}\langle S_i^z\rangle + 2\gamma_0\langle S_i^+S_i^-\rangle + 2\gamma_0(N-1)\{\langle S_i^+S_i^-\rangle - \langle S_i^+S_j^+S_i^-S_j^-\rangle\} = 0 \ , \tag{3.2}$$

and hence the total energy (in units of ω_0) satisfies the equation

$$\frac{\partial W}{\partial t} + 2\gamma_0(W+\tfrac{1}{2}N) + 2\gamma_0(N-1)\{W+\tfrac{1}{2}N - N\langle S_i^+S_j^+S_i^-S_j^-\rangle\} = 0 \ . \tag{3.3}$$

The radiation rate $I(t)$ is then given by

$$I(t) = -\omega_0\frac{\partial W}{\partial t} = 2\gamma_0\omega_0\{N(W+\tfrac{1}{2}N) - N(N-1)\langle S_i^+S_j^+S_i^-S_j^-\rangle\} \ . \tag{3.4}$$

It is seen from (3.3) and (3.4) that the radiation rate can be determined if the two-particle mean value $\langle S_i^+S_j^+S_i^-S_j^-\rangle$ is known. On the other hand, it is easily seen from Eq. (1.1) that the two-particle mean value $\langle S_i^+S_j^+S_i^-S_j^-\rangle$ satisfies the following equation[14]:

$$\frac{\partial}{\partial t}\langle S_i^+S_j^+S_i^-S_j^-\rangle + 4\gamma_0\langle S_i^+S_j^+S_i^-S_j^-\rangle + \gamma_0\sum_{l\neq i\neq j}\{\langle S_i^+S_j^+S_j^-S_l^-\rangle + \langle S_i^+S_j^+S_i^-S_l^-\rangle + \text{c.c.}\} = 0 \quad (i\neq j) \ . \tag{3.5}$$

We thus obtain the whole hierarchy of equations. We now describe various decoupling procedures. The nature of the decoupling will, of course, depend on the initial excitation of the system. We consider two types of excitation.

A. Atomic System Initially Excited to State (2.6) with $\theta_0 < \pi$

In the case of an atomic system initially excited to state (2.6) with $\theta < \pi$, we proceed, as was also done in I, by making a "Hartree-type" approximation on the two-particle mean value

$$\langle S_i^+S_j^+S_i^-S_j^-\rangle = \langle S_i^+S_i^-\rangle\langle S_j^+S_j^-\rangle \quad (i\neq j) \ . \tag{3.6}$$

On combining (3.6) and (3.4) we obtain a simple equation for $\langle S_i^+S_i^-\rangle$, which is easily solved subject to the initial condition $\langle S_i^+S_i^-\rangle = (\frac{1}{2} - \frac{1}{2}\cos\theta_0)$. The radiation rate is then found to be given by the well-known[1,12] "sech" solution

$$I(t) = \frac{\omega_0\gamma_0N^3}{2(N-1)}\operatorname{sech}^2\{N\gamma_0(t-\tau)\} \ , \qquad \tau = \frac{1}{2N\gamma_0}\ln\frac{(N-1)}{1+N\cot^2\frac{1}{2}\theta_0} \ . \tag{3.7}$$

It is now desirable to test the accuracy of the Hartree-type approximation (3.6). Now in general it is well known[15] that the inaccuracy introduced would be smaller the higher the order of the mean values for which the decoupling is done. With this in mind, we now make a Hartree-type approximation on the three-particle mean value:

$$\langle S_i^+S_j^+S_k^+S_k^-\rangle = \langle S_i^+S_j^-\rangle\langle S_k^+S_k^-\rangle \quad (i\neq j\neq k) \ . \tag{3.8}$$

It is easily verified that a Hartree-Fock-type approximation will be inconsistent with the initial condition. On substituting (3.8) in (3.5) we obtain

$$\frac{\partial}{\partial t}\langle S_i^+S_j^+S_i^-S_j^-\rangle + 4\gamma_0\langle S_i^+S_j^+S_i^-S_j^-\rangle + (N-2)\gamma_0\langle S_j^+S_j^-\rangle \times(\langle S_i^+S_l^-\rangle + \text{c.c.}) + (N-2)\gamma_0\langle S_i^+S_i^-\rangle(\langle S_j^+S_l^-\rangle + \text{c.c.}) = 0 \ , \tag{3.9}$$

where in writing (3.9) we have made use of the permutation symmetry. We multiply Eq. (3.1) by $\langle S_j^+S_j^-\rangle$ and add to it the corresponding equation obtained by replacing i by j. We then obtain the following equation:

$$\frac{\partial}{\partial t}\{\langle S_i^+S_i^-\rangle\langle S_j^+S_j^-\rangle\} + 4\gamma_0\langle S_i^+S_i^-\rangle\langle S_j^+S_j^-\rangle + (N-1)\gamma_0\langle S_j^+S_j^-\rangle \times(\langle S_i^+S_l^-\rangle + \text{c.c.}) + (N-1)\gamma_0\langle S_i^+S_i^-\rangle \times(\langle S_j^+S_l^-\rangle + \text{c.c.}) = 0 \ . \tag{3.10}$$

On combining (3.9) and (3.10), we obtain the following relation:

$$[\langle S_i^+S_j^+S_i^-S_j^-\rangle - \langle S_i^+S_i^-\rangle\langle S_j^+S_j^-\rangle(N-2)/(N-1)]e^{4\gamma_0t} = \text{const} \ . \tag{3.11}$$

The constant of integration is found to be equal to $[1/(N-1)]\sin^4\frac{1}{2}\theta_0$ on using the initial condition. This result leads us to conclude that the fluctuation

$\langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle$ is of the order of $1/N$ and therefore the inaccuracy introduced by the approximation (3.6) is quite small for large N values. From (3.11), we also have

$$\langle S_i^+ S_j^+ S_i^- S_j^- \rangle = \frac{N-2}{(N-1)N^2}(W+\tfrac{1}{2}N)^2 + \frac{\sin^4\frac{1}{2}\theta_0}{N-1}\, e^{-4\gamma_0 t} . \tag{3.12}$$

On substituting (3.12) in (3.3) we obtain the equation

$$\frac{\partial W}{\partial t} + 2\gamma_0 N(W+\tfrac{1}{2}N) - 2\gamma_0(1-2/N)(W+\tfrac{1}{2}N)^2 - 2\gamma_0 N \sin^4\tfrac{1}{2}\theta_0\, e^{-4\gamma_0 t} = 0 . \tag{3.13}$$

It should be noted that this equation is in the form of the Riccati equation[10] and can be solved by standard techniques. Its solution, which is given in Appendix A, is

$$W(t) = \frac{N}{N-2}\left[1 - \frac{d}{d\tau}\ln f(\tau)\right], \quad \tau = 2\gamma_0 t \tag{3.14}$$

where

$$f(\tau) = x[J'_{N/2}(x)Y_{N/2}(xe^{-\tau}) - Y'_{N/2}(x)J_{N/2}(xe^{-\tau})] + \beta[J_{N/2}(xe^{-\tau})Y_{N/2}(x) - Y_{N/2}(xe^{-\tau})J_{N/2}(x)] \tag{3.15}$$

and

$$x = \sin^2\tfrac{1}{2}\theta_0 (N-2)^{1/2}, \quad \beta = -[1 + (\tfrac{1}{2}N - 1)\cos\theta_0] . \tag{3.16}$$

Here J_ν and Y_ν are the Bessel functions of first and second kind, respectively. Equation (3.14) can now be used to obtain an approximation to the estimate of the radiation rate that is better than the one given by (3.7).

B. Atomic System Initially Excited to the State $\rho(0) = \Pi_i |+\rangle_{i\,i}\langle +|$

In the case of an atomic system initially excited to the state $\rho(0) = \Pi_i |+\rangle_{i\,i}\langle +|$, the dipole moment of the system remains zero and we must make an approximation which takes into account the correlations between different atoms. In this case the approximation (3.6) is inadequate; however, an Hartree-Fock-type approximation turns out to be a good one. This approximation is

$$\langle S_i^+ S_j^+ S_i^- S_j^- \rangle = \langle S_i^+ S_i^- \rangle \langle S_j^+ S_j^- \rangle + \langle S_i^+ S_j^- \rangle \langle S_j^+ S_i^- \rangle \quad (i \neq j) . \tag{3.17}$$

On using the relation $\langle \vec{S}_i \cdot \vec{S}_j \rangle = \frac{1}{4}$, Eq. (3.17) may be written as

$$\langle S_i^+ S_j^+ S_i^- S_j^- \rangle = \langle S_i^+ S_i^- \rangle \langle S_j^+ S_j^- \rangle + (\langle S_i^+ S_i^- \rangle - \langle S_i^+ S_j^+ S_i^- S_j^- \rangle)^2 ,$$

which leads to

$$\langle S_i^+ S_j^+ S_i^- S_j^- \rangle = \langle S_i^+ S_i^- \rangle + \tfrac{1}{2} - \{\tfrac{1}{4} + \langle S_i^+ S_i^- \rangle - \langle S_i^+ S_i^- \rangle^2\}^{1/2} . \tag{3.18}$$

On substituting (3.18) in (3.3), we obtain the following equation for W:

$$\frac{dW}{dt} + 2\gamma_0(W+\tfrac{1}{2}N) - \gamma_0 N(N-1) + 2\gamma_0 N(N-1)(\tfrac{1}{2} - W^2/N^2)^{1/2} = 0 . \tag{3.19}$$

The solution of (3.19) is reduced to quadratures and is found to be given by

$$-\frac{b}{b^2+c^2}\ln(a + b\cos x + c\sin x) + \frac{c}{b^2+c^2}\, x - \frac{ac}{b^2+c^2}(b^2+c^2-a^2)^{-1/2}\ln\left|\frac{(a-b)\tan\frac{1}{2}x + c - (b^2+c^2-a^2)^{1/2}}{(a-b)\tan\frac{1}{2}x + c + (b^2+c^2-a^2)^{1/2}}\right| = 2\gamma_0(t-t_0) , \tag{3.20}$$

where

$$a = -N + 2/\sqrt{2}, \quad b = 1, \quad c = (N-1), \quad W = (N/\sqrt{2})\cos x . \tag{3.21}$$

In Eq. (3.20), t_0 is determined from the initial condition $x(0) = \frac{1}{4}\pi$. For large values of N, expression (3.20) is considerably simplified and we have

$$\left|\frac{\tan\frac{1}{2}x - \alpha}{\tan\frac{1}{2}x - \beta}\right| = \exp[2N\gamma_0(t-t_0) - x] , \tag{3.22}$$

where

$$\alpha = \sqrt{2} - 1 - (2\sqrt{2}/N)(\sqrt{2}-1), \quad \beta = (\sqrt{2}+1) . \tag{3.23}$$

Equations (3.22) and (3.21) enable us to calculate the time dependence of the energy of the system and the radiation rate is then given by

$$I(t) = 2\gamma_0\omega_0[W + N - \tfrac{1}{2}N^2 + (N-1)(\tfrac{1}{2}N^2 - W^2)^{1/2}] . \tag{3.24}$$

Our results (3.7) and (3.24) for the radiation rate are found to be in good agreement with the results obtained by the direct numerical integration[6] of the master equation (1.2).

IV. NEW DESCRIPTION OF SPONTANEOUS EMISSION

In this section we obtain a different type of Fokker-Planck equation which describes spontaneous emission. We will show that the solution to the problem of spontaneous emission from two-level atoms is equivalent to the solution of $2N$ coupled nonlinear equations which provide a new insight into this problem. We introduce the characteristic function[16] $C(\{\alpha_i\}, \{\alpha_i^*\}, t)$ defined by

$$C(\{\alpha_i\},\{\alpha_i^*\},t)\equiv\langle\prod_i e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}\rangle\ . \tag{4.1}$$

It is obvious that this characteristic function is useful in computing the expectation values of the form $\langle S_i^+S_j^+S_k^+\cdots S_l^-S_m^-\cdots\rangle$. The desired equation of motion for C can be obtained by combining (1.1) and (4.1):

$$\frac{\partial C}{\partial t}=-\gamma_0\sum_i \mathrm{Tr}\left((S_i^+S_i^-\rho-2S_i^-\rho S_i^++\rho S_i^+S_i^-)\prod_k e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}\right)-\gamma_0\sum\sum_{i\neq j}\mathrm{Tr}\left((S_i^+S_j^-\rho-2S_j^-\rho S_i^++\rho S_i^+S_j^-)\prod_k e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}\right), \tag{4.2}$$

which can be rewritten as

$$\frac{\partial C}{\partial t}=-\gamma_0\sum_i\left\langle\prod_{k\neq i}e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}(e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^+S_i^- - 2S_i^+e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^- + S_i^+S_i^-e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-})\right\rangle$$

$$-\gamma_0\sum\sum_{i\neq j}\left\langle\prod_{k\neq i\neq j}e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}(e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^+e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-}S_j^-$$

$$-2S_i^+e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-}S_j^- + S_i^+e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_j^-e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-})\right\rangle. \tag{4.3}$$

To simplify Eq. (4.3) we make use of the following identities[17]:

$$e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^+S_i^- = e^{i\alpha_i S_i^+}S_i^+e^{i\alpha_i^* S_i^-}S_i^- + (i\alpha_i^*)e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^-\ , \tag{4.4}$$

$$e^{i\alpha_i^* S_i^-}S_i^+ = S_i^+e^{i\alpha_i^* S_i^-} - 2i\alpha_i^*S_i^+S_i^-e^{i\alpha_i^* S_i^-} + i\alpha_i^*e^{i\alpha_i^* S_i^-} - (i\alpha_i^*)^2S_i^-e^{i\alpha_i^* S_i^-}\ , \tag{4.5}$$

and their Hermitian adjoints. Then (4.3) reduces to

$$\frac{\partial C}{\partial t}=-\gamma_0\sum_i\left\langle\prod_{k\neq i}e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}(i\alpha_i^*e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}S_i^- + i\alpha_i e^{i\alpha_i S_i^+}S_i^+e^{i\alpha_i^* S_i^-})\right\rangle-\gamma_0\sum\sum_{i\neq j}\left\langle\prod_{k\neq i\neq j}e^{i\alpha_k S_k^+}e^{i\alpha_k^* S_k^-}\right.$$

$$\times\{[-2i\alpha_i^*S_i^+e^{i\alpha_i S_i^+}S_i^-e^{i\alpha_i^* S_i^-} + i\alpha_i^*e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-} - (i\alpha_i^*)^2e^{i\alpha_i S_i^+}S_i^-e^{i\alpha_i^* S_i^-}]e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-}S_j^- + S_i^+e^{i\alpha_i S_i^+}e^{i\alpha_i^* S_i^-}$$

$$\left.\times[-2i\alpha_j e^{i\alpha_j S_j^+}S_j^+S_j^-e^{i\alpha_j^* S_j^-} + i\alpha_j e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-} - (i\alpha_j)^2e^{i\alpha_j S_j^+}e^{i\alpha_j^* S_j^-}S_j^-]\}\right\rangle\ . \tag{4.6}$$

It should now be noted that various expectation values appearing on the right-hand side of (4.6) can be expressed in terms of the derivatives of $C(\{\alpha_i\},\{\alpha_i^*\},t)$. Then (4.6) reduces to

$$\frac{\partial C}{\partial t}=-\gamma_0\sum\sum_{ij}\left[(i\alpha_i^*)\frac{\partial C}{\partial(i\alpha_j^*)}+(i\alpha_i)\frac{\partial C}{\partial(i\alpha_j)}\right]$$

$$+\gamma_0\sum\sum_{i\neq j}\left[(i\alpha_i)^2\frac{\partial^2 C}{\partial(i\alpha_i)\partial(i\alpha_j)}+(2i\alpha_i)\frac{\partial^3 C}{\partial(i\alpha_i)\partial(i\alpha_i^*)\partial(i\alpha_j)}+(i\alpha_i^*)^2\frac{\partial^2 C}{\partial(i\alpha_i^*)\partial(i\alpha_j^*)}+(2i\alpha_i^*)\frac{\partial^3 C}{\partial(i\alpha_i)\partial(i\alpha_i^*)\partial(i\alpha_j^*)}\right]. \tag{4.7}$$

Since the operators $S_i^\pm$ satisfy the relation $(S_i^\pm)^2=0$, Eq. (4.7) is equivalent to the equation

$$\frac{\partial C}{\partial t}=-\gamma_0\sum\sum_{ij}\left[(i\alpha_i^*)\frac{\partial C}{\partial(i\alpha_j^*)}+(i\alpha_i)\frac{\partial C}{\partial(i\alpha_j)}\right]+2\gamma_0\sum\sum_{i\neq j}\left[(2i\alpha_i^*)\frac{\partial^3 C}{\partial(i\alpha_i)\partial(i\alpha_i^*)\partial(i\alpha_j^*)}+(2i\alpha_i)\frac{\partial^3 C}{\partial(i\alpha_i)\partial(i\alpha_i^*)\partial(i\alpha_j)}\right]. \tag{4.8}$$

We now introduce the distribution function[16] $P(\{z_i\},\{z_i^*\},t)$ defined by

$$P(\{z_i\},\{z_i^*\},t)=\frac{1}{\pi^{2N}}\int d^2\{\alpha_i\}C(\{\alpha_i\},\{\alpha_i^*\},t)\prod_i e^{-i(\alpha_i z_i^*+\alpha_i^* z_i)}. \tag{4.9}$$

On substituting (4.8) in (4.9) we can obtain the time dependence of P, which is given by

$$\frac{\partial P}{\partial t}=\gamma_0\sum_{ij}\sum\left[\frac{\partial}{\partial z_i}(z_j P)+\frac{\partial}{\partial z_i^*}(z_j^* P)\right]-2\gamma_0\sum_{i\neq j}\sum\left[\frac{\partial}{\partial z_i}(|z_i|^2 z_j P)+\frac{\partial}{\partial z_i^*}(|z_i|^2 z_j^* P)\right]. \tag{4.10}$$

The Fokker-Planck equation (4.10) is equivalent to the following Langevin[18] equations:

$$\dot{z}_i=-\gamma_0\sum_j z_j+2\gamma_0|z_i|^2\sum_{j\neq i}z_j,\quad i=1,2,\ldots,N. \tag{4.11}$$

These are the Langevin equations which provide an alternative description of spontaneous emission from a system of N two-level atoms and a solution to these equations should provide us with the entire dynamics of the atoms emitting spontaneously. It is also of interest to note that (4.11) is a kind of equation of motion for an N-dimensional van der Pol oscillator[19] and equations of similar type also occur in the description of multimode lasers.[20] We first note that the steady-state solution of (4.11) corresponds to $z_i=0$, which leads to the following for the steady-state solution of the Fokker-Planck equation:

$$P_{\text{steady}}(\{z_i\},\{z_i^*\})=\prod_i\delta^{(2)}(z_i). \tag{4.12}$$

On combining (4.12) and (4.9) we find that

$$C_{\text{steady}}(\{\alpha_i\},\{\alpha_i^*\})=1\ \text{ for all } \{\alpha_i\} \text{ and } \{\alpha_i^*\}. \tag{4.13}$$

It is obvious then that

$$\rho_{\text{steady}}=\prod_i|-\rangle_{ii}\langle-|, \tag{4.14}$$

as expected. We recall that the Langevin equations which described spontaneous emission from a system of harmonic oscillators were[1,2] [cf. Eq. (II 2.3)]

$$\dot{z}_i=-\gamma_0\sum_j z_j. \tag{4.15}$$

On comparing (4.11) with (4.15) we clearly see the type of nonlinearity which occurs for the problem of spontaneous emission from two-level atoms. For *one two-level* atom, there is of course no nonlinearity and one has the solution $z(t)=z(0)\times e^{-\gamma_0 t}$, which can be shown to lead to all the well-known results for spontaneous emission from one two-level atom.

The Langevin equations (4.11) are very useful in obtaining the equations of motion for the mean values: It is clear from (4.11) that $|z_i|^2$ satisfies the equation of motion

$$\frac{d}{dt}(|z_i|^2)=-\gamma_0\sum_j(z_i^* z_j+z_i z_j^*), \tag{4.16}$$

where we have made use of the *auxiliary constraint* $z_i^2=z_i^{*2}=0$. On taking the expectation value of (4.16), we obtain the equation

$$\frac{d}{dt}\langle S_i^+ S_i^-\rangle=-\gamma_0\sum_j(\langle S_i^+ S_j^-\rangle+\langle S_j^+ S_j^-\rangle), \tag{4.17}$$

which is easily seen to lead to Eq. (3.1). Similarly, from the Langevin equations (4.11) we obtain the following equation for $(|z_i|^2|z_j|^2)$ $(i\neq j)$:

$$\frac{d}{dt}(|z_i|^2|z_j|^2)=-4\gamma_0|z_i|^2|z_j|^2-\gamma_0\sum_{k\neq i\neq j}(z_k z_i^*|z_j|^2+z_k z_j^*|z_i|^2+\text{c.c.}). \tag{4.18}$$

On taking the mean value, Eq. (4.18) reduces to

$$\frac{d}{dt}\langle S_i^+ S_j^+ S_i^- S_j^-\rangle=-4\gamma_0\langle S_i^+ S_j^+ S_i^- S_j^-\rangle-\gamma_0\sum_{k\neq i\neq j}(\langle S_i^+ S_j^+ S_j^- S_k^-\rangle+\langle S_j^+ S_i^+ S_i^- S_k^-\rangle+\text{c.c.}), \tag{4.19}$$

which is the desired (3.5).

V. SPONTANEOUS EMISSION FROM TWO-LEVEL ATOMS WITH INHOMOGENEOUS BROADENING

In our treatment[1,2] of the spontaneous emission from identical N two-level atoms, and also in other publications,[5,12,13] it was assumed that all the atoms had the same energy separation between the two levels. In this section we consider the effects of inhomogeneous broadening,[21] i.e., we assume that the jth atom has the frequency ω_j, the atoms being identical otherwise. Such a circumstance is highly desirable because substances, such as ruby, which are usually employed for experiments have finite inhomogeneous broadening. Moreover there are phenomena such as photon echoes[22] for which the presence of inhomogeneous broadening is crucial. A general theory should, therefore, take into account the effects of inhomogeneous broadening and this will be done now. The interaction Hamiltonian between N two-level atoms and the quantized field is now equal to

$$H=\sum_j\omega_j S_j^Z+\sum_{ks}\omega_{ks}a_{ks}^\dagger a_{ks}+\sum_{ks}\sum_j(S_j^+ a_{ks}g_{jks}+\text{H.c.}), \tag{5.1}$$

where the coupling constant g_{jks} is given by [compare it with that given by Eq. (I 2.2)]

$$g_{jks}=\frac{-i\omega_j}{c}\left(\frac{2\pi c}{L^3}\right)^{1/2}\frac{1}{\sqrt{k}}(\vec{\epsilon}_{ks}\cdot\vec{d}). \tag{5.2}$$

Starting with the Hamiltonian (5.1), we can obtain the master equation for the reduced density operator $\rho(t)$, in the interaction picture, corresponding

to the atomic system. The details are given in Appendix B, where we find that the approximate master equation is

$$\frac{\partial\rho}{\partial t} = -\gamma_0 \sum_{j1} (S_j^+ S_1^- \rho - S_1^- \rho S_j^+) e^{i(\omega_j-\omega_1)t} + \text{H. c.} \tag{5.3}$$

In the derivation of (5.3), we assumed that the frequencies ω_j are clustered around some central frequency ω_0 and that $|\omega_j - \omega_0| < \omega_0$. Under this condition we can replace g_{jks} by g_{ks}, where g_{ks} is obtained from (5.2) by the replacement $\omega_j = \omega_0$. Equation (5.3) leads to the following equation for the mean value $\langle S_i^z(t)\rangle$:

$$\frac{\partial}{\partial t}\langle S_i^z(t)\rangle = -\gamma_0 \sum_i \{\langle S_i^+ S_i^-\rangle e^{i(\omega_i-\omega_i)t} + \text{c. c.}\} . \tag{5.4}$$

The radiation rate is then given by

$$I(t) = -\frac{d}{dt}\sum_i \omega_i \langle S_i^z(t)\rangle \approx -\omega_0 \frac{d}{dt}\sum_i \langle S_i^z(t)\rangle$$

$$= 2\gamma_0 \omega_0 \sum_{ij} (\langle S_i^+ S_j^-\rangle e^{i(\omega_i-\omega_j)t}) . \tag{5.5}$$

Thus to obtain the radiation rate, $\langle S_i^+ S_j^-\rangle$ has to be calculated. This expectation value is, in turn, to be obtained by the solution of the master equation (5.3). We will again assume that the system was initially excited to the state (2.6). We make the Hartree-type approximation $\langle S_i^+ S_j^+ S_i^- S_j^-\rangle = \langle S_i^+ S_i^-\rangle \langle S_j^+ S_j^-\rangle$ and moreover assume that even though there is inhomogeneity in the medium the expectation values of the form $\langle S_i^z\rangle$ and $\langle S_i^+ S_j^-\rangle$, where operators refer to the interaction picture, are roughly same for any pair of atoms. We think that this will be well satisfied because of the condition $|\omega_j - \omega_0| \ll \omega_0$. From (5.4) we obtain the equation

$$\frac{dW}{dt} = \frac{d}{dt}\sum_i \langle S_i^z\rangle = -\gamma_0 \left(\sum_{ij} \langle S_i^+ S_j^-\rangle e^{i(\omega_i-\omega_j)t} + \text{c. c.}\right) . \tag{5.6}$$

Because of the assumed validity of the relation $\langle \vec{S}_i \cdot \vec{S}_j\rangle = \frac{1}{4}$, Eq. (5.6) can be written in the form

$$\frac{dW}{dt} = -2\gamma_0(W + \tfrac{1}{2}N) - 2\gamma_0(\tfrac{1}{4} - W^2/N^2) \sum_{i\neq j} (e^{i(\omega_i-\omega_j)t}) , \tag{5.7}$$

where we have also assumed that initially each atom was similarly excited. Let us introduce the quantity $\Gamma(t)$ defined by

$$\Gamma(t) = \left|\textstyle\sum_i e^{i\omega_i t}/N\right|^2 . \tag{5.8}$$

Equation (5.7) becomes

$$\frac{dW}{dt} = -2\gamma_0(W + \tfrac{1}{2}N) - 2\gamma_0(\tfrac{1}{4} - W^2/N^2)[N^2\Gamma(t) - N] . \tag{5.9}$$

The solution of (5.9) subject to the initial condition $W(0) = -\frac{1}{2}N\cos\theta_0$ is given by

$$W(t) = -\tfrac{1}{2}N + N[2\gamma_0\psi(t) + 1 + \cot^2\tfrac{1}{2}\theta_0\chi(t)]^{-1} , \tag{5.10}$$

where

$$\chi(t) = \exp(2N\gamma_0 \textstyle\int_0^t \Gamma(\tau)d\tau), \quad \psi(t) = \chi(t)\int_0^t d\tau\chi^{-1}(\tau) . \tag{5.11}$$

On using (5.10), we obtain the following expression for the radiation rate:

$$I(t) = 2\gamma_0\omega_0[1 + 2N\gamma_0\Gamma(t)\psi(t) + N\Gamma(t)\cot^2\tfrac{1}{2}\theta_0\chi(t)]$$

$$\times[1 + 2\gamma_0\psi(t) + \cot^2\tfrac{1}{2}\theta_0\chi(t)]^{-2} . \tag{5.12}$$

It is seen from (5.12) that the radiation rate is a functional of $\Gamma(t)$, which is *equal to the modulus square of the Fourier transform of the atomic-line-shape factor.*

One may carry out a similar analysis for the case when each two-level atom is replaced by a harmonic oscillator. It is found that the Fokker-Planck equation describing the spontaneous emission from this system is

$$\frac{\partial F_s^{(A)}}{\partial t} = \gamma_0 \sum_{j1} \frac{\partial}{\partial z_j}[z_1 F_s^{(A)}] e^{i(\omega_j-\omega_1)t} + \text{c. c.} \tag{5.13}$$

The notation is the same as that of II. We assume that at time $t = 0$, each of the oscillators was excited to a coherent state $|z_0\rangle$; the solution of (5.13) is then

$$F_s^{(A)}(\{z_i\}, \{z_i^*\}, t) = \textstyle\prod_i [\pi\delta^{(2)}(z_i - \langle z_i(t)\rangle)] , \tag{5.14}$$

where $\langle z_i(t)\rangle$ is the solution of the equation

$$\langle \dot{z}_i(t)\rangle = -\gamma_0 \textstyle\sum_j e^{i(\omega_i-\omega_j)t}\langle z_j(t)\rangle , \tag{5.15}$$

subject to the initial condition $\langle z_j(0)\rangle = z_0$. We again assume that $\langle z_i(t)\rangle$ is approximately independent of the index i; the solution of the Eq. (5.15) is then

$$\langle z_i(t)\rangle \approx z_0 \exp[-N\gamma_0 \textstyle\int_0^t dt_1 \Gamma(t_1)] , \tag{5.16}$$

where $\Gamma(t)$ is defined as before. The energy of the system is equal to

$$W(t) = \textstyle\sum_j \omega_j \langle a_j^\dagger a_j\rangle \approx \omega_0 \sum_j |\langle z_j(t)\rangle|^2$$

$$= \omega_0 N |z_0|^2 \exp[-2N\gamma_0 \textstyle\int_0^t dt_1 \Gamma(t_1)] . \tag{5.17}$$

From (5.17), we then obtain the following expression for the radiation rate:

$$I(t) = 2N^2 |z_0|^2 \omega_0\gamma_0 \Gamma(t) \exp[-2N\gamma_0 \textstyle\int_0^t dt_1 \Gamma(t_1)] . \tag{5.18}$$

This formula exhibits explicitly the effects of inhomogeneous broadening on the intensity of the spontaneously emitted radiation.

APPENDIX A: SOLUTION OF EQ. (3.13)

In this appendix, we will present the solution of

Eq. (3.13). On introducing the parameter $\tau = 2\gamma_0 t$, we can write (3.13) in the form

$$\frac{d\varphi}{d\tau} = -N\varphi + (1-2/N)\varphi^2 + N\sin^4\tfrac{1}{2}\theta_0 e^{-2\tau}, \quad \varphi = (W+\tfrac{1}{2}N)\ . \tag{A1}$$

It should be noted that the Eq. (A1) is a Riccati-type equation and can be solved by using the standard techniques.[10] We first make the transformation

$$\varphi = -\left(\frac{N}{N-2}\right)\frac{d}{d\tau}\ln\chi\ . \tag{A2}$$

Then χ is found to satisfy the following second-order differential equation:

$$\frac{d^2\chi}{d\tau^2} + N\frac{d\chi}{d\tau} + (N-2)\sin^4\tfrac{1}{2}\theta_0 e^{-2\tau}\chi = 0\ . \tag{A3}$$

On changing the independent variable τ to y defined by

$$y = \tfrac{1}{4}(N-2)\sin^4\tfrac{1}{2}\theta_0 e^{-2\tau}\ , \tag{A4}$$

we obtain the equation

$$y\frac{d^2\chi}{dy^2} + (1-\tfrac{1}{2}N)\frac{d\chi}{dy} + \chi = 0\ . \tag{A5}$$

The solution to (A5) can be shown to be

$$\chi(y) = y^{N/4}\left[A J_{N/2}(2\sqrt{y}) + B Y_{N/2}(2\sqrt{y})\right], \tag{A6}$$

where $J_{N/2}$ and $Y_{N/2}$ are Bessel functions of the first and second kind, respectively. The constants A and B are to be determined from the initial condition. On introducing the original variable τ, we have

$$\chi(\tau) = (\tfrac{1}{2}x)^{N/2}e^{-N\tau/2}\left[A J_{N/2}(xe^{-\tau}) + B Y_{N/2}(xe^{-\tau})\right], \quad x = (N-2)^{1/2}\sin^2\tfrac{1}{2}\theta_0\ . \tag{A7}$$

On substituting (A7) in (A2), we obtain, after some simplifications,

$$\varphi(\tau) = \frac{N^2}{2(N-2)} + \frac{Nxe^{-\tau}}{N-2}\,\frac{J'_{N/2}(xe^{-\tau}) + (B/A)Y'_{N/2}(xe^{-\tau})}{J_{N/2}(xe^{-\tau}) + (B/A)Y_{N/2}(xe^{-\tau})}\ , \tag{A8}$$

where prime indicates the derivative with respect to the argument. On using the initial condition $\varphi(0) = \frac{1}{2}N(1-\cos\theta_0)$, we obtain the following expression for the ratio B/A:

$$\frac{B}{A} = \frac{J'_{N/2}(x) - (\beta/x)J_{N/2}(x)}{(\beta/x)Y_{N/2}(x) - Y'_{N/2}(x)}\ , \quad \beta = -\left[1 + (\tfrac{1}{2}N-1)\cos\theta_0\right]. \tag{A9}$$

On substituting (A9) in (A8) and making use of the relation $W(\tau) = \varphi(\tau) - \frac{1}{2}N$, we obtain after a number of simplifications, the results (3.14) and (3.15) given in the text.

APPENDIX B: DERIVATION OF MASTER EQUATION (5.3) AND (5.13)

We first derive the master equation (5.13) for the case of N harmonic oscillators emitting spontaneously. We will use the general approach developed in Ref. 3. The desired master equation in the Born approximation is given by Eq. (G4.36), viz.,

$$\frac{\partial F_s^{(A)}}{\partial t} + \int_0^t K(t,\tau)d\tau = 0\ , \tag{B1}$$

where the kernel is given by Eq. (G 4.37a). For the problem under consideration, the correlation matrix, as given by Eq. (G 4.37b) is

$$\Gamma^{ks,k's'}(t,\tau) = \begin{pmatrix} 0 & e^{-i\omega_{ks}(t-\tau)} \\ 0 & 0 \end{pmatrix}\delta_{kk'}\delta_{ss'}\ . \tag{B2}$$

The kernel (G 4.37a) then simplifies to

$$K(t,\tau) = \sum_{ks} e^{-i\omega_{ks}(t-\tau)}\{\psi^*_{ks}(t)(\mathfrak{D}_s^{(1)} - \mathfrak{D}_s^{(2)}) \times[\psi_{ks}(\tau)\,\mathfrak{D}_s^{(1)}F_s^{(A)}(\tau)]\} + \text{c.c.} \tag{B3}$$

Here the differential operators $\mathfrak{D}_s^{(1)}$ and $\mathfrak{D}_s^{(2)}$ [cf. Eq. (G 4.35)] for the antinormal rule of mapping are given by[16(a),(b)]

$$\mathfrak{D}_s^{(1)} = \exp\left(-\sum_j \frac{\overleftarrow{\partial}}{\partial z_j^*}\frac{\overrightarrow{\partial}}{\partial z_j}\right), \quad \mathfrak{D}_s^{(2)} = \exp\left(-\sum_j \frac{\overleftarrow{\partial}}{\partial z_j}\frac{\overrightarrow{\partial}}{\partial z_j^*}\right), \tag{B4}$$

and the system functions $\psi_{ks}(t)$ are given by

$$\psi_{ks}(t) = \textstyle\sum_j g_{jks}z_j e^{-i\omega_j t}\ . \tag{B5}$$

We now assume that the inhomogeneity is such that $g_{jks} \approx g_{ks}$, where g_{ks} is obtained from (5.2) by the substitution $\omega_j = \omega_0$. On substituting (B3)–(B5) in (B1), we obtain the following master equation:

$$\frac{\partial F_s^{(A)}}{\partial t} = \gamma_0\sum_{ij}\frac{\partial}{\partial z_i}\left[z_j F_s^{(A)}\right]e^{i(\omega_i-\omega_j)t} + \text{c.c.} \tag{B6}$$

In the derivation of (B6), we took the infinite volume limit and made use of the Markovian approximation. Equation (B6) is the master equation which has the effects of inhomogeneous broadening in it. The corresponding equation of motion for the reduced density operator may be obtained by the use of the identities[23]

$$\Omega^{(A)}\left(\frac{\partial}{\partial z_i}(z_j F_s^{(A)})\right) = -\left[a_i^\dagger, a_j\rho_s\right], \tag{B7a}$$

$$\Omega^{(A)}\left(\frac{\partial}{\partial z_i^*}(z_j^* F_s^{(A)})\right) = +\left[a_i, \rho_s a_j^\dagger\right], \tag{B7b}$$

where $\Omega^{(A)}$ is the mapping operator[16(a),(b)] for the antinormal rule of association. The master equation for the reduced density operator is

$$\frac{\partial\rho_s}{\partial t} = -\gamma_0\sum_{ij}\left[a_i^\dagger, a_j\rho_s\right]e^{i(\omega_i-\omega_j)t} + \text{H.c.} \tag{B8}$$

The derivation of the master equation for the case of two-level atoms is algebraically more involved

and will not be presented here. The derivation is exactly similar to the one given above and the result (which can also be obtained formally by replacing a_j by S_j^- and $a_j^\dagger$ by S_j^+) is

$$\frac{\partial \rho_s}{\partial t} = -\gamma_0 \sum_{ij} [S_i^+, S_j^- \rho_s] e^{i(\omega_i - \omega_j)t} + \text{H.c.}\,, \tag{B9}$$

which is Eq. (5.3).

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[1]G. S. Agarwal, Phys. Rev. A 2, 2038 (1970). This paper will be referred to as I. All equations from this paper will be prefixed by I.

[2]G. S. Agarwal, Phys. Rev. A 3, 1783 (1971). This paper will be referred to as II. All equations from this paper will be prefixed by II.

[3]G. S. Agarwal, Phys. Rev. 178, 2025 (1969) and references therein. Equations of this reference will be preceded by the letter G.

[4]R. P. Feynman, F. L. Vernon, Jr., and R. W. Hellwarth, J. Appl. Phys. 28, 49 (1957).

[5]R. H. Dicke, Phys. Rev. 93, 99 (1954).

[6]A master equation similar to (1.2) has also been derived by R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302 (1971). They, however, use a different model than ours.

[7]This is a general feature of many-body systems. For a discussion of various approximate schemes used in classical statistical mechanics, see the classical paper of N. N. Bogoliubov, in *Studies in Statistical Mechanics*, edited by J. DeBoer and G. E. Uhlenbeck (North-Holland, Amsterdam, 1962), Vol. I; see in particular Secs. 6 and 7.

[8]This result has been summarized in G. S. Agarwal, Opt. Commun. 2, 357 (1971).

[9]See, for example, H. Haken, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1970), Vol. XXV/2C.

[10]For an excellent discussion of various types of nonlinear equations see, for example, H. T. Davis, *Introduction to Nonlinear Differential and Integral Equations* (Dover, New York, 1960).

[11]E. T. Jaynes and F. W. Cummings, Proc. IEEE 51, 89 (1963); C. R. Willis, J. Math. Phys. 5, 1241 (1964).

[12]J. H. Eberly and N. Rehler, Phys. Letters 29A, 142 (1969); Phys. Rev. A 3, 1735 (1971). There are, however, some differences in the time dependence of $\theta(t)$ as given by (2.16) and that quoted by the above authors.

[13](a) R. H. Lehmberg, Phys. Rev. A 2, 883 (1970); (b) D. Dialetis, *ibid.* 2, 599 (1970); (c) J. H. Eberly and N. Rehler, *ibid.* 2, 1607 (1970).

[14]Equations (3.1) and (3.5) for the mean values are also derived in Sec. IV by using the Langevin equations which describe the spontaneous emission from two-level atoms.

[15]See, for example, D. N. Zubarev, Usp. Fiz. Nauk. 71, 71 (1960) [Sov. Phys. Usp. 3, 320 (1960)].

[16]For a detailed discussion of the characteristic function and the associated phase-space distribution functions, see (a) G. S. Agarwal and E. Wolf, Phys. Rev. D 2, 2161 (1970); (b) 2, 2187 (1970); (c) M. Lax, Phys. Rev. 172, 350 (1968); (d) Ref. 9, p. 65.

[17]These relations are easily established by the repeated use of the following identities: $[A, e^{-xB}] = -\int_0^x dy\, e^{-(x-y)B} \times [A,B] e^{-yB}$ and $e^{xB} A\, e^{-xB} = A + x[B,A] + (x^2/2!)[B,[B,A]] + \cdots$, which are valid for any two operators A and B. For a discussion of these identities, see, e.g., R. M. Wilcox, J. Math. Phys. 8, 962 (1967). Identities similar to (4.4) and (4.5) of the present paper are also given in Ref. 9, p. 69.

[18]For a detailed discussion of the equivalence of the Fokker-Planck equations to Langevin equations, see, for example, R. L. Stratonovich, *Topics in the Theory of Random Noise* (Gordon and Breach, New York, 1963), Vol. I, or M. Lax, Rev. Mod. Phys. 38, 541 (1966).

[19]B. van der Pol, Phil. Mag. 3, 65 (1927); Proc. IRE 22, 1051 (1934).

[20]See, for example, Ref. 9, p. 169.

[21]This problem has been independently investigated by J. H. Eberly, Nuovo Cimento Letters 1, 182 (1971); Acta. Phys. Polon. (to be published). In the latter paper a result similar to (5.5), but not to (5.12), has been derived on the basis of the theory developed in Ref. 12. The author is pleased to thank Dr. Eberly for a discussion of the problem of inhomogeneous broadening.

[22]I. D. Abella, N. A. Kurnit, and S. R. Hartman, Phys. Rev. 141, 391 (1966). In Paper IV of this series, we hope to discuss a dynamical theory of photon echoes.

[23]These identities are easily established by using the general theory developed in Ref. 16(a).