

## Spontaneous emission from a single two-level atom in the presence of $N$ initially unexcited identical atoms

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The quantum-electrodynamical problem of  $N$  identical two-level atoms interacting with  $M$  field modes is considered, and an exact solution for the case of spontaneous emission from a single atom is obtained. The behavior of this system is shown to be quite different from the exponential decay of a single excited atom radiating into free space. A "ringing" behavior at the enigmatic frequency  $\sim\sqrt{N}$  which occurs when only one mode is accessible to the field persists when many modes are present. When a continuum of modes is accessible, the long-time limit shows that the energy of the originally excited atom is equally likely to be shared by the  $N-1$  originally unexcited atoms and the electromagnetic field when  $M/N \rightarrow 1$ . "Radiation suppression" occurs whenever  $N \gg 1$  and  $M \ll N$ , when the originally excited atom does not emit its energy. When  $M = N \gg 1$ , the single atom decays monotonically to zero (and approximately exponentially) but with a decay rate that is  $\sqrt{N}$  times the Rabi frequency. Such unexpected effects are presently within experimental range.

### INTRODUCTION

The present work presents the time variation of a quantum-electrodynamical system, a radiation field in interaction with an atomic (or nuclear) system. Recent experimental advances have brought such time-dependent solutions into testable range, and will provide a different kind of comparison of theory and experiment than the usual perturbation calculation and related experimental determination of energy levels, such as in the well-known "Lamb shift."

Dicke<sup>1</sup> first emphasized the cooperative nature of the spontaneous emission from a system of identical atoms where the atoms were at equivalent mode positions, e.g., located within a space of linear dimension small compared to a wavelength. He showed that an assemblage of identical two-level atoms, if prepared in a certain way, can exhibit "superradiance," or spontaneous emission which is proportional to the square of the number of radiating atoms. The present paper presents an exact solution for the spontaneous emission of a single atom which is initially excited in the presence of  $N-1$  initially unexcited identical but distinguishable atoms, and where there are  $M$  modes of the electromagnetic field accessible to the radiation. The solution is exact in the sense that the long-time behavior of the average number of photons in a given mode is given in closed form for arbitrarily large field-atom couplings in terms of elementary functions under the conditions that the atoms ( $N \gg 1$ ) are at random space positions. Such solutions valid for all times for  $N$ -body quantum systems are invariably interesting in their own right because of their expository and pedagogic value. The present model is further interesting in that it couples two different quantum systems,  $N$  atoms and  $M$  field modes. The problem of  $N$  two-level atoms with one atom initially excited emitting spontaneously into a single accessible electromagnetic mode has previously been given

exactly,<sup>2</sup> when the phenomenon of "radiation suppression"<sup>3</sup> is clarified, and energy is exchanged between field and atoms with the enigmatic frequency of the square root of the number of atoms  $N$  times the one-atom frequency. The atom effectively will not emit its energy as the number of atoms  $N$  becomes very large, and the energy is trapped in the single atom. Whether these intriguing phenomena persist when there are  $M$  modes of the field present with frequencies near the atomic resonance will be a question of focus in this paper. Further, it is well known that a single (isolated) excited atom radiating into free space will decay (approximately) exponentially to the ground state,<sup>4,5</sup> and it will be of interest to see how this result is modified when there are many other atoms present [cf. below Eq. (35) and Fig. 4].

Recently there have been significant advances in cold cavity techniques with Rydberg atoms<sup>6-9</sup> which bring a number of previously inaccessible theoretical predictions (see Haroche<sup>10</sup> for a review) within the purview of the experimentalist, for example the time dependence of a single atom in a single mode of the electromagnetic field.<sup>6</sup> The existence of huge electric dipole matrix elements between nearby levels of Rydberg atoms makes the intrinsic single-atom transition frequency unusually large, and further, the resonant frequencies between these levels fall in the millimeter range, corresponding to large size low-order cavities in which it is easy to prepare and keep atoms in a region of well defined and constant atom-field coupling. Rydberg atoms usually have large spontaneous emission even in free space, ensuring that their coupling to the other modes of the field can be neglected during their interaction with the selected mode of the cavity. This means that they constitute in these experiments quasi-ideal two-level systems in which all the levels corresponding to the nonresonant transitions of the cavity are irrelevant.

A recent experiment of Pavolini *et al.*<sup>11</sup> with gallium

atoms gives convincing evidence of "subradiance," when spontaneous emission is quenched by destructive interatomic interference similar to that predicted by Dicke<sup>1</sup> when two atoms are prepared in an antisymmetrical state. The present paper gives a theoretical calculation of spontaneous emission in the presence of  $N \gg 1$  atoms which is valid for arbitrarily large atom-field coupling and arbitrary times, and it is now reasonable to hope that features of this system will come under experimental scrutiny in the near future. One example of a prediction is the persistence of "ringing" of the total photon number even when the number of modes becomes large, an effect which does not occur when one atom alone radiates into a large number of modes, when the field build up is monotonic and closely exponential. The origin of the "suppression" which is predicted here appears to be different from that seen by Pavolini *et al.*,<sup>11</sup> and thus constitutes a different effect, as the present quenching depends only on  $N \gg 1$  and not on a coherent antisymmetrical initial preparation of atomic state.

### MATHEMATICAL FORMULATION

The mathematical setup of the problem has been often discussed before.<sup>1,5,12</sup> The Hamiltonian for the problem is given by ( $\hbar = 1$ )

$$H = \sum_{\mu=1}^M \omega_{\mu} a_{\mu}^{\dagger} a_{\mu} + \Omega \sum_{j=1}^N \sigma_j^z + \left[ \sum_{\mu,j} \lambda_{\mu j} \sigma_j^{(-)} a_{\mu}^{\dagger} + \text{H.c.} \right]. \quad (1)$$

The operator  $a_{\mu}$  is the photon destruction operator for the  $\mu$ th mode, and  $\sigma_j^{(-)}$  is a "step-down" operator for the  $j$ th atom. The operators satisfy the commutator relations

$$[a_{\mu}, a_{\mu'}^{\dagger}] = \delta_{\mu\mu'}, \quad (2)$$

and

$$[\sigma_j^{(+)}, \sigma_{j'}^{(-)}] = 2\sigma_j^z \delta_{jj'}. \quad (3)$$

The  $\lambda_{\mu j}$  are defined as

$$\lambda_{\mu j} = \boldsymbol{\mu} \cdot \mathbf{E}_{\mu}^{(+)}(\mathbf{x}_j) / 2, \quad (4)$$

where  $\boldsymbol{\mu}$  is the electric (magnetic) dipole moment, and  $E_{\mu}^{(+)}$  is the (positive) space part of the electromagnetic field at the position of the two-level atom. If  $H$  is written as  $H_0 + H_1$ , where  $H_1 = 0$  for  $\lambda = 0$ , then the matrix elements of  $H$  can be expressed in a basis of eigenstates of  $H_0$ .

Attention is focused on the situation where the initial energy corresponds to the presence of zero photons in the field, and  $N - 1$  of the atoms unexcited, so that there is one unit of energy  $\Omega$  available to be shared among the parts of the system. How is this energy transferred in time? There are  $N + M$  states which span the subspace of interest. The base states, eigenstates of  $H_0$ , can be defined as follows:

$$|\mu\rangle = |-, -, \dots, -, 1_{\mu}\rangle \quad (4a)$$

(one photon is present in mode  $\mu$  and all atoms are unexcited; there are  $M$  such states), and

$$|j\rangle = |-, -, \dots, +, -, \dots; 0_{\mu}\rangle \quad (4b)$$

(one atom, the  $j$ th, is excited, all others unexcited, and there are no photons in the field; there are  $N$  such states).

The only matrix elements are

$$H_{\mu\mu'} = (\omega_{\mu} - \Omega) \delta_{\mu\mu'} \equiv 2\Delta_{\mu} \delta_{\mu\mu'}, \quad (5)$$

and

$$H_{\mu j} = \lambda_{\mu j}, \quad H_{j j'} = 0.$$

A constant  $\Omega$  times the unit matrix has been subtracted from  $H$  with no loss of generality, so that the elements  $H_{j j'}$  are all zero.

The density operator has the form (Schrödinger representation)

$$\rho(t) = \exp(iHt) \rho(0) \exp(-iHt). \quad (6)$$

Interest attaches to the diagonal matrix elements  $\rho_{\mu\mu} \equiv n_{\mu}(t)$ , the probability that a photon has been emitted into mode  $\mu$  at time  $t$ , and to  $\rho_{j j}$ , the probability that atom  $j$  is excited. A calculation of the  $\mu j$ th element of  $H$  is also required. In what follows, the symbol  $H_{\mu j}^n$  will be used to denote the  $\mu j$ th matrix element of  $H^n$ , the parentheses being deleted for brevity. This is

$$H_{\mu j}^n = \sum_{\mu'} H_{\mu\mu'} H_{\mu' j}^{n-1} + \sum_j H_{\mu j} H_{j j}^{n-1}, \quad (7)$$

and

$$H_{j j}^{n-1} = \sum_{\mu} H_{j \mu} H_{\mu j}^{n-2} = \sum_{\mu} \lambda_{\mu j}^* H_{\mu j}^{n-2}. \quad (8)$$

Thus (7) can be written as, using Eq. (5),

$$H_{\mu j}^n = 2\Delta_{\mu} H_{\mu j}^{n-1} + \sum_{\mu', j'} \lambda_{\mu j} \lambda_{\mu' j'}^* H_{\mu' j'}^{n-2}. \quad (9)$$

At this point, use will be made of the reasonable assumption that the atoms are at random space positions  $\mathbf{x}_j$ , so that

$$\sum_j \lambda_{\mu j} \lambda_{\mu' j}^* = \Lambda_{\mu}^2 \delta_{\mu\mu'}, \quad (10)$$

$$\Lambda_{\mu}^2 = \sum_j |\lambda_{\mu j}|^2. \quad (11)$$

This is most easily seen to be appropriate in the case of free-space modes, when

$$\sum_j \lambda_{\mu j} \lambda_{\mu' j}^* = \sum_j \Lambda_{\mu} \Lambda_{\mu'} \exp[i(\mathbf{k}_{\mu} - \mathbf{k}_{\mu'}) \cdot \mathbf{x}_j]. \quad (12)$$

With this assumption, Eq. (9) takes the form

$$H_{\mu j}^n = 2\Delta_{\mu} H_{\mu j}^{n-1} + \Lambda_{\mu}^2 H_{\mu j}^{n-2}. \quad (13)$$

The difference equation (13) has the solution, in view of the values  $H_{\mu j}^1 = \lambda_{\mu j}$ , and  $H_{\mu j}^2 = 2\Delta_{\mu} \lambda_{\mu j}$ ,

$$H_{\mu j}^n = \lambda_{\mu j} (h_+^n - h_-^n) / 2\Gamma_{\mu}, \quad (14)$$

where

$$h_{\pm} = \Delta_{\mu} \pm \Gamma_{\mu}, \quad (15)$$

$$\Gamma_{\mu} = (\Delta_{\mu}^2 + \Lambda_{\mu}^2)^{1/2}. \quad (16)$$

The exponential in the expression for the density matrix can be summed to give for the probability of finding a photon in mode  $\mu$ ,

$$\begin{aligned} \rho_{\mu\mu} &= \sum_{a,b} [\exp(iHt)]_{\mu a} \rho_{ab}(0) [\exp(-iHt)]_{b\mu} \\ &= |[\exp(iHt)]_{\mu j}|^2, \end{aligned} \quad (17)$$

since  $\rho_{ab}(0) = \delta_{ab} \delta_{aj}$ , where atom  $j$  was the initially excited one. Use of Eq. (14) gives

$$\begin{aligned} \rho_{\mu\mu}(t) = n_{\mu}(t) &= \left| \sum_{n=0}^{\infty} \frac{(it)^n}{n!} \frac{\lambda_{\mu j}}{2\Gamma_{\mu}} (h_+^n - h_-^n) \right|^2 \\ &= |\lambda_{\mu j}|^2 \sin^2 \Gamma_{\mu} t / \Gamma_{\mu}^2. \end{aligned} \quad (18)$$

For the case that only one mode is accessible to the radiation, Eq. (18) reduces to the expression, in the special case that all atoms are in equivalent mode positions, so that the  $\lambda_{\mu j}$  are all equal,

$$n(t) = \sin^2(\sqrt{N} \lambda t) / N, \quad \left. \begin{array}{l} \Delta_{\mu} = 0, \\ \lambda_{\mu j} = \lambda \end{array} \right\}. \quad (19)$$

For  $N$  very large, no photons are emitted, and the energy remains trapped in the  $j$ th atom.<sup>2,3</sup> Here the frequency  $\sqrt{N} \lambda$  appears as the frequency of transfer of energy between the field and atomic system. Unlike the explanation given by Kaluzny *et al.*<sup>9</sup> in their observation of the ringing regime of superradiance when an ensemble of  $N$  atoms radiates in the cavity a field whose amplitude is  $\sqrt{N}$  times larger than a single-photon field, thus increasing the Rabi-exchange frequency to  $\sqrt{N} \lambda$ , the origin of the present ringing frequency, although of the same magnitude, is not at all apparent since it results when only one photon is present (at most) in the field.

It is pertinent to emphasize the differences between the present effect, here dubbed radiation suppression, and the quite different phenomenon of Dicke's subradiance.<sup>1</sup> Recall that Dicke's effect arises from a particular preparation of a system of  $N$  identical two-level systems in an antisymmetrical state. It is helpful to think of the simplest case of two neutron spins in a dc magnetic field, with one spin initially excited and the other unexcited. The two are confined to a volume small compared to a wavelength of any emitted radiation  $E = hc/\lambda$ , where  $E$  is a neutron level spacing  $E = \mu H_0$ . The singlet state corresponds pictorially to an initial preparation of state such that the perpendicular components of the individual spins precess out of phase, and thus the net magnetic moment is zero. Since the radiation rate (by Fermi's "golden rule") is proportional to the square of the magnetic (electric) dipole moment, the system of the two neutrons so prepared does not transit to the state of both being unexcited. Conversely, when the two spins are prepared initially in phase, the dipole moment  $\sim \sqrt{2}\mu$  (or more generally  $[N/2(N/2+1)]^{1/2}\mu$ ) and the rate is  $2I_0$ , where  $I_0$  is the isolated atom radiation rate; this latter is termed superradiance by Dicke, when the rate is in general  $N/2(N/2+1)I_0$ .

In contrast to subradiance, the radiation suppression

described here does not depend on an initial preparation of appropriate phases of dipole moments; the atoms are here also required to be at disparate mode positions, so that the sum over atoms gives a delta function in mode index [cf. Eq. (10)], in the case of many modes  $M$ . All that is further required is that all atoms except one be initially unexcited. Both of the somewhat mysterious effects of radiation suppression and oscillations at a frequency  $\sim \sqrt{N} \lambda$  are thus of a different nature than Dicke's subradiance.

The total probability that a photon has been emitted into any mode is the trace over all modes of the elements

$$n(t) = \sum_{\mu} n_{\mu}(t) = \sum_{\mu} |\lambda_{\mu j}|^2 \sin^2(\Gamma_{\mu} t) / \Gamma_{\mu}^2. \quad (20)$$

This expression will be examined in the case that  $\Lambda_{\mu}^2 = \lambda^2 N$ , where  $N$  is the effective number of atoms coupled to the field, and  $\lambda_{\mu}^2$  is independent of  $\mu$ . Taking the sum over to the integral gives

$$n(t) = \int_{-x_0}^{x_0} \rho(x) \{ \sin^2 [T(x^2 + N)^{1/2}] / (x^2 + N) \} dx, \quad (21)$$

where the dimensionless variables  $T = \lambda t$ ,  $x = \Delta/\lambda$  are defined, and the number of modes  $M$  is

$$\int \rho dx = M. \quad (22)$$

In the case that  $\rho$  can be taken as a constant over the range of interest, then the long-time limit of Eq. (21), obtained by expressing the squared sine term in terms of the double-angle expression and dropping the time-dependent term, is easily seen to be

$$n(t) \rightarrow (M/2N) [\sqrt{N}/x_0 \tan^{-1}(x_0/\sqrt{N})], \quad (23)$$

and is thus approximately  $M/2N$  when  $x_0$  is small compared to  $\sqrt{N}$ .

When  $x_0$  is small compared to  $\sqrt{N}$ , the integral may be well approximated by expanding the square root and keeping the  $x^2$  term, and neglecting the  $x$  dependence in the denominator. This leads to the expression

$$n(t) \simeq (M/2N) \{ 1 - A(t) \cos[2\sqrt{N} \lambda t + \varphi(t)] \}, \quad (24)$$

where the slowly varying amplitude function  $A$  is unity for  $t=0$  and approaches zero as  $(\sqrt{N}/t)^{1/2}$  when  $t$  goes to infinity. It is given in terms of the well-known Fresnel integrals  $C$  and  $S$  by

$$A(z) = \{ [C^2(z) + S^2(z)] / z^2 \}^{1/2}, \quad (25)$$

where

$$C(z) = \int_0^z \cos(\pi s^2/2) ds, \quad (26)$$

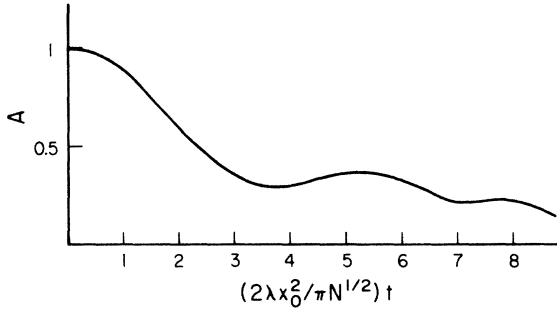
$$S(z) = \int_0^z \sin(\pi s^2/2) ds, \quad (27)$$

and

$$z^2 = 2\lambda t x_0^2 / \pi \sqrt{N}. \quad (28)$$

$A$  is shown in Fig. 1 as a function of the variable  $z^2 \sim t$ . The amplitude  $A$  falls to zero as  $t^{-1/2}$ , since the integrals  $C$  and  $S$  approach  $\frac{1}{2}$  as  $t$  approaches infinity.

A calculation similar to the one just given, and involving only knowledge of  $H_{\mu j}^n$ , allows calculation of  $\rho_{jj}$ , the

FIG. 1. Amplitude  $A$  versus time.

probability of finding the initially excited atom  $j$  in the upper state, since  $H_{jj}^n$  is simply the mode sum of  $H_{\mu j}^{n-1}$ , given by

$$H_{jj}^n = \sum_{\mu} H_{j\mu} H_{\mu j}^{n-1} = \sum_{\mu} \lambda_{\mu j}^* H_{\mu j}^{n-1}. \quad (29)$$

This leads to the expression for the probability of excitation of the  $j$ th atom of

$$\rho_{jj}(t) = \left| \sum_{\mu} \frac{|\lambda_{\mu j}|^2}{2\Gamma_{\mu}} \left[ \frac{\exp(ih_+ t)}{h_+} - \frac{\exp(ih_- t)}{h_-} - \left( \frac{1}{h_+} - \frac{1}{h_-} \right) \right] + 1 \right|^2, \quad (30)$$

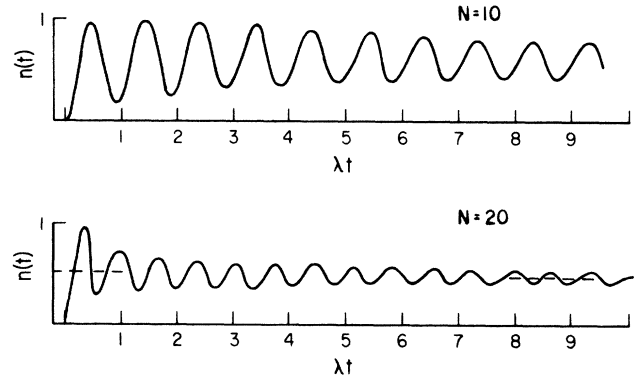
where  $h_{\pm}$  and  $\Gamma_{\mu}$  are defined by Eqs. (15) and (16). The long time limit of this expression, in the case that all couplings are taken equal,  $\lambda_{\mu j} = \lambda$ , is

$$\rho_{jj}(t) \rightarrow \left[ 1 - \sum_{\mu} |\lambda_{\mu j}|^2 / \Lambda_{\mu}^2 \right]^2 \rightarrow (1 - M/N)^2 \quad (31)$$

as  $t \rightarrow \infty$ . The probability that the initially unexcited atoms are excited, taken as a whole, is given simply as unity minus (the probability that a photon has been emitted plus the probability that the atom  $j$  has been deexcited); that is to say

$$\sum_{\substack{j, j' \\ j' \neq j}} \rho_{j'j'} = 1 - \rho_{jj}(t) - n(t) \rightarrow 1 - (1 - M/N)^2 - M/2N \quad (32)$$

as  $t \rightarrow \infty$ . The conclusion is that radiation suppression does not persist as the number of accessible modes  $M$  ap-

FIG. 2. Total photon number  $n$  as a function of  $\lambda t$  for two values of  $N$ , and for  $x_0 = \Delta_0/\lambda \gg \sqrt{N}$ .

proaches the number of atoms  $N$ , as can be seen from Eqs. (31) or (32); when  $M=N$ , half of the energy goes into the field and the other half is transferred to the initially unexcited atoms. This limit is similar to the phenomenon of classical radiation trapping.<sup>3</sup>

Numerical integration of Eq. (21) for the probability that there are  $n$  photons in the field in the case that the density of modes is assumed constant over a frequency band  $x_0 \gg \sqrt{N}$  shows that the oscillatory (or ringing) behavior seen in the case of  $x_0 \ll \sqrt{N}$  (see Fig. 1) persists. Figure 2 shows the situation of a broad spread of mode frequencies for two values of the total atom number  $N$  and for which  $x_0 \gg \sqrt{N}$ . Figure 2 shows a behavior which does not resemble an exponential buildup. Also seen plotted in Fig. 3 is  $n(t)$  as a function of  $\sqrt{N} \lambda t$  for the case of  $M=N$  and  $x_0 = \sqrt{N}$ . As mentioned before, spontaneous emission into many modes by an isolated atom shows an exponential field buildup, and the ringing is thus purely a feature of the  $N$  atom nature.

In the case that  $\Gamma_{\mu} = \sqrt{N} \lambda (x^2 + 1)^{1/2}$  and  $x = \Delta/\sqrt{N} \lambda$ , the expression for the probability of finding the  $j$ th atom excited at time  $t$  [Eq. (30)] may be written as

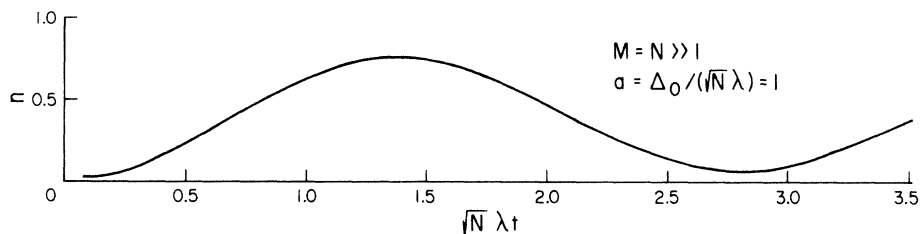
$$\rho_{jj}(t) = (1 + I_1)^2 + I_2^2, \quad (33)$$

where

$$I_1 = \left[ \frac{M}{2Na} \right] \int_{-a}^a \frac{\cos\{[x + (x^2 + 1)^{1/2}]T\} - 1}{(x^2 + 1)^{1/2}[x + (x^2 + 1)^{1/2}]} dx, \quad (34)$$

and

$$I_2 = \left[ \frac{M}{2Na} \right] \int_{-a}^a \frac{\sin\{[x + (x^2 + 1)^{1/2}]T\}}{(x^2 + 1)^{1/2}[x + (x^2 + 1)^{1/2}]} dx. \quad (35)$$

FIG. 3. Total photon number  $n$  as a function of  $\sqrt{N} \lambda t$  for  $M=N \gg 1$ , and  $\Delta_0 = \sqrt{N} \lambda$ .

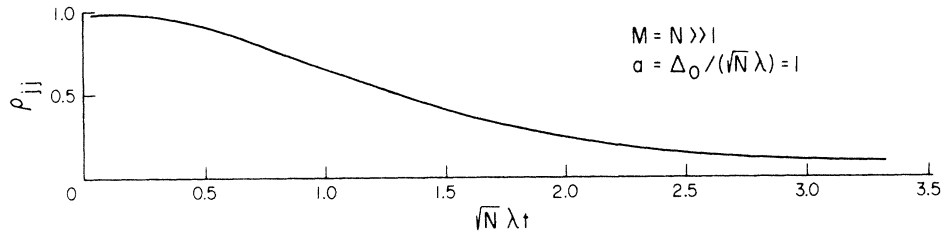


FIG. 4.  $\rho_{jj}$ , the probability of finding atom  $j$  excited as a function of  $\sqrt{N}\lambda t$ .

Here  $T \equiv \sqrt{N}\lambda t$  and  $a = \Delta_0 / \sqrt{N}\lambda$ , and the density of states has again been taken as a constant over the range of interest. Figure 4 shows  $\rho_{jj}(t)$  as a function of  $\sqrt{N}\lambda T$ , and for  $M = N \gg 1$  and  $a = 1$ . It is seen from this figure that the decay of the initially excited atom to its ground state is, after a short "induction" time, roughly exponential in the variable  $\sqrt{N}\lambda t$ . Thus the decay rate is proportional to  $\sqrt{N}$  (as is the ringing frequency of the field and of the  $N - 1$  atoms), a quite different result than for an isolated atom radiating into free space.

No interesting new features are obtained by assuming various continuous mode profiles which have a single peak at resonance for  $\rho(x)$  which are not already illustrated by the constant density cases. The case of bimodal or

other more complicated mode profiles have not been investigated, however.

The case of free-space corresponding to an infinite number of modes in an infinite volume must be accompanied by a limit of infinite atom number  $N$ , since otherwise the energy, or correspondingly the trace of the density matrix, will not be preserved, as it is apparent that  $n$ , the photon number, must always be less than or equal to unity. Thus an apparent divergence of the integral of Eq. (21) when a free-space density of modes is assumed is avoided by simultaneously letting  $N$  go to infinity. An abbreviated version of this work has appeared previously.<sup>13</sup>

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