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Intensity Fluctuations in Many-Atom Spontaneous Emission

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The normalized two-time intensity-fluctuation functions are calculated for the spontaneous-emission radiation from a system of N two-level atoms. The radiated field, when the system decays from the excited state to the ground state, does not look like a Gaussian or a coherent field.

Statistical properties of the radiation field emitted into free space by a system of N identical two-level atoms (the dimensions of which are much smaller than the radiation wavelength) have been recently studied[1,2] by means of the single-time second-order normalized correlation function $g^{(2)}$. When the system is in the excited state, $g^{(2)}$ roughly equals 2. This value is typical of the Brown and Twiss effect[3] for thermal or pseudothermal Gaussian fields. When the system decays from the excited state to the superradiant state,[4] the behavior of $g^{(2)}$ is essentially classical: $g^{(2)}$ decreases to nearly 1, which is the value for a coherent field. But, as the system decays further to the ground state, the intensity-fluctuation function calculated in a model in which the radiation field is quantized is quite different from the classical one: larger and larger correlations appear as the system is drawn near the ground state.[2] We can say that the radiation field becomes more and more *incoherent*[5] as the atoms approach the ground state. In this paper, we propose to explicate the nature of the spontaneous emission radiation from a many-atom system by means of the two-time intensity-fluctuation function.

The two-time normalized second-order correlation function is defined as

$$g^{(2)}(r_1, t_1; r_0, t_0) = \frac{\langle A^-(r_0, t_0)A^-(r_1, t_1)A^+(r_1, t_1)A^+(r_0, t_0)\rangle}{\langle A^-(r_0, t_0)A^+(r_0, t_0)\rangle\langle A^-(r_1, t_1)A^+(r_1, t_1)\rangle} . \quad (1)$$

It can be shown, by using the Heisenberg equations of motion for the photon annihilation operators, that the positive frequency part $\vec{A}^+(r, t)$ of the potential vector is the sum of the free-potential vector operator and of a term proportional to the retarded atomic lowering operator $R_-(t - |r|/c)$.[4] (The system is located at $r = 0$.) Hence, in terms of the system operators, the normalized intensity-fluctuation function is given by

$$g^{(2)}(\tau, \tau_0) = \frac{\langle R_+(\tau_0)R_+(\tau+\tau_0)R_-(\tau+\tau_0)R_-(\tau_0)\rangle}{\langle R_+(\tau_0)R_-(\tau_0)\rangle\langle R_+(\tau_0+\tau)R_-(\tau_0+\tau)\rangle} . \quad (2)$$

In this equation, τ_0 is the retarded time and $\tau = t_1 - t_0 - (|r_1| - |r_0|)/c$ is the delay time, chosen positive or zero.

Under the Markoff approximation, the quantization of the radiation field leads to the equation[6]

$$\frac{d\rho}{dt} = -\frac{1}{2}(R_+R_-\rho + R_+R_-\rho - 2R_-\rho R_+) = \Lambda\rho \quad (3)$$

for the reduced-density matrix of the system, with t in units of the radiation lifetime of an atom. The solution of Eq. (3) is

$$\rho(t) = e^{\Lambda(t-t_0)}\rho(t_0) . \quad (4)$$

With the help of the quantum regression theorem,[7] the intensity fluctuations

$$G^{(2)}(\tau, \tau_0) = \langle R_+(\tau_0)R_+(\tau_0+\tau)R_-(\tau_0+\tau)R_-(\tau_0)\rangle \quad (5)$$

can be written by using the operator Λ defined by Eq. (3) as[8]

$$G^{(2)}(\tau, \tau_0) = \mathrm{Tr}R_+R_-e^{\Lambda\tau}[R_-\rho(\tau_0)R_+] . \quad (6)$$

Then, if we define

$$\sigma(\tau, \tau_0) = e^{\Lambda\tau}[R_-\rho(\tau_0)R_+] , \quad (7)$$

the normalized intensity-fluctuation function can be written

$$g^{(2)}(\tau, \tau_0) = \frac{\sum \nu_m \sigma_{m,m}(\tau, \tau_0)}{\sum \nu_m \rho_{m,m}(\tau_0) \sum \nu_m \rho_{m,m}(\tau+\tau_0)} . \quad (8)$$

In this equation $\nu_m = (\frac{1}{2}N + m)(\frac{1}{2}N - m + 1)$ with $m = -\frac{1}{2}N, \ldots, \frac{1}{2}N$. As, by its definition, $\sigma(\tau, \tau_0)$ obeys the same equation as $\rho(t)$ [Eq. (3)], we can calculate $g^{(2)}(\tau, \tau_0)$ by numerical integration of the matrix elements $\rho_{m,m}(\tau)$ and $\sigma_{m,m}(\tau, \tau_0)$, with

$$\sigma_{m,m}(0, \tau_0) = \nu_{m+1}\rho_{m+1,m+1}(\tau_0) . \quad (9)$$

Let us derive a relation which will be useful to discuss our numerical results. Consider the difference

$$\langle R_+(t_1)R_+(t_2)R_-(t_2)R_-(t_1)\rangle - \langle R_+(t_2)R_-(t_2)R_+(t_1)R_-(t_1)\rangle$$
$$= \mathrm{Tr} R_+R_-e^{\Lambda(t_2-t_1)}[R_-\rho(t_1)R_+ - R_+R_-\rho(t_1)]. \quad (10)$$

From Eq. (3), we find that

$$\langle R_+(t_1)R_+(t_2)R_-(t_2)R_-(t_1)\rangle - \langle R_+(t_1)R_-(t_1)R_+(t_2)R_-(t_2)\rangle$$
$$= \mathrm{Tr} R_+R_-e^{\Lambda(t_2-t_1)}\left(\frac{d}{dt}\rho(t)\right)_{t=t_1} = \left(\frac{d}{dt}\langle R_+R_-\rangle\right)_{t=t_2} \quad (11)$$

This relation is of some interest, when at time t_1 the system is in a pure state. Then Eq. (11) becomes

$$\left(\frac{d}{dt}\langle R_+R_-\rangle\right)_{t=t_2} = \langle R_+(t_1)R_+(t_2)R_-(t_2)R_-(t_1)\rangle - \langle R_+(t_2)R_-(t_2)\rangle\langle R_+(t_1)R_-(t_1)\rangle. \quad (12)$$

Then, the normalized intensity-fluctuation function is

$$g^{(2)}(\tau,\tau_0) = 1 + \frac{1}{\langle R_+(\tau_0)R_-(\tau_0)\rangle} \times \frac{d}{d\tau}\ln\langle R_+(\tau+\tau_0)R_-(\tau+\tau_0)\rangle. \quad (13)$$

Let us first consider the system in the excited state at time $\tau_0 = 0$. Figure 1 shows $g^{(2)}(\tau,\tau_0)$ as a function of delay $N\tau$ for different values of the system's normalized energy $W(\tau_0)/N$, where we have chosen $N = 20$. In the plane $N\tau = 0$, we have reported Fig. 1 of Ref. 2.

For times τ_0 so that $W(\tau_0) > 0$, $g^{(2)}(\tau,\tau_0)$ monotonically decreases from its initial value to zero. But, as the system decays further to the ground state, $g^{(2)}(\tau,\tau_0)$ begins to increase from $g^{(2)}(0,\tau_0)$ when $N\tau$ goes up [see Fig. 1 for $W(\tau_0)/N < -\frac{1}{4}$]. To confirm our numerical calculations we can calculate the $g^{(2)}(\tau,\tau_0)$ derivative at $\tau = 0$. With the help of the Eqs. (3) and (7)–(9), we find

$$\frac{d}{d\tau}g^{(2)}(\tau,\tau_0)_{\tau=0} = \frac{2}{\langle R^+(\tau_0)R^-(\tau_0)\rangle^3}\sum_{m>k}\nu_m\nu_k(m-k) \times [\nu_{m+1}\rho_{k,k}\rho_{m+1,m+1} - \nu_{k+1}\rho_{k+1,k+1}\rho_{m,m}]. \quad (14)$$

The $\rho_{m,m}$'s distribution given by Eq. (3) is a smooth function of m, and monotonic near the ground or excited state (see Fig. 4 of Ref. 2). In these two cases, we can approximate Eq. (14) by

$$\left(\frac{d}{d\tau}g^{(2)}(\tau,\tau_0)\right)_{\tau=0} \approx \frac{2}{\langle R_+(\tau_0)R_-(\tau_0)\rangle^3}\sum_{m>k}\nu_m\nu_k(m-k)\rho_{k,k}\rho_{m,m}(\nu_{m+1}-\nu_{k+1})$$
$$\approx -\frac{2}{\langle R_+(\tau_0)R_-(\tau_0)\rangle^3}\sum_{m>k}\nu_m\nu_k(m-k)^2(m+k+1)\rho_{k,k}\rho_{m,m}. \quad (15)$$

So, one clearly sees that the $g^{(2)}(\tau,\tau_0)$ derivative at $\tau = 0$ is negative when the populated states have a positive energy, while it becomes positive as the system approaches the ground state. However, as the system decays further in such a manner that the only populated states are $|-\frac{1}{2}N+2\rangle$, $|-\frac{1}{2}N+1\rangle$, and $|-\frac{1}{2}N\rangle$, the $g^{(2)}(\tau,\tau_0)$ derivative at $\tau = 0$ becomes again negative [Eq. (14)]. For a two-atom system, one finds that $g^{(2)}(\tau,\tau_0) = g^{(2)}(0,\tau_0)(1+2\tau_0)/[1+2(\tau_0+\tau)]$ is a decreasing function of the time τ.

To illustrate the behavior of $g^{(2)}(\tau,\tau_0)$ in the region of negative energy, we refer to Fig. 2. τ_0 is chosen so that $W(\tau_0)/N$ is equal to -0.4. For $N = 4$ one has the situation described by the third case of Eq. (14) discussed. For larger N, $g^{(2)}(\tau,\tau_0)$ reaches a maximum before decreasing to zero. It is not apparent how to explain why the fluctuations are maxima for a time delay different from zero. We can only presume that the radiation field is not Gaussian because $g^{(2)}(\tau,\tau_0)$ is not maximum for zero time delay.

For $\tau_0 = 0$, as the system is known to emit incoherently, we can compare the exact $g^{(2)}(\tau,0)$ given by Eq. (13) with the expression

$$g^{(2)}_{\mathrm{inc}}(\tau,0) = 1 + |g^{(1)}(\tau,0)|^2, \quad (16)$$

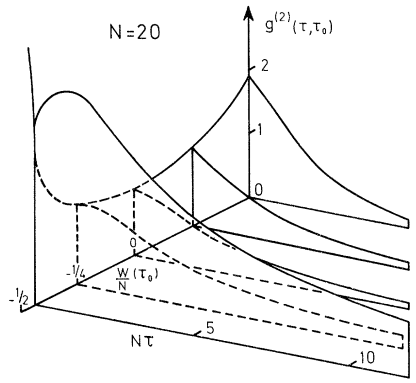

FIG. 1. Normalized intensity-fluctuation function $g^{(2)}(\tau,\tau_0)$ vs atomic energy $W(\tau_0)/N$ and retarded time delay $N\tau$ for $N = 20$.

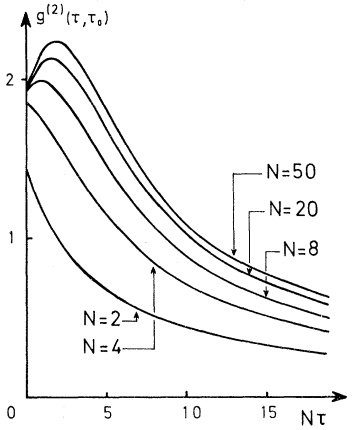

FIG. 2. Normalized intensity-fluctuation function $g^{(2)}(\tau,\tau_0)$ vs retarded time delay $N\tau$ with $W(\tau_0)/N=-0.4$, for different values of N.

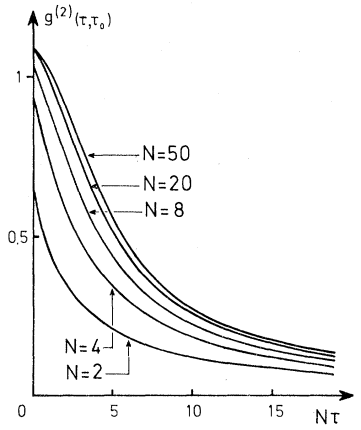

FIG. 4. Normalized intensity-fluctuation function $g^{(2)}(\tau,\tau_0)$ vs $N\tau$ with $W(\tau_0)=0$ for different N.

where $g^{(1)}(\tau,0)$ is the two-time first-order normalized correlation function. The relation (16) expresses that the photoelectron counts obey the Bose-Einstein statistics.

The function $g^{(1)}(\tau,0)$ has been calculated in the same way as $g^{(2)}(\tau,0)$. In Fig. 3, the full line is the exact result and the broken line corresponds to Eq. (16) for $N=20$. One immediately concludes that the radiation field is not Gaussian. Even for very short delays, the photoelectron counts do not obey the Bose-Einstein statistics. Indeed the derivatives of $g^{(2)}(\tau,0)$ and $g^{(2)}_{\rm inc}(\tau,0)$ at time $\tau=0$ are found to be

$$\left(\frac{d}{d\tau}\,g^{(2)}(\tau,0)\right)_{\tau=0}=-4+\frac{4}{N}\,, \tag{17}$$

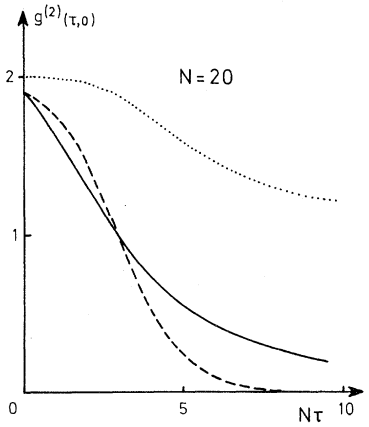

FIG. 3. Normalized intensity-fluctuation function $g^{(2)}(\tau,0)$ with $W(0)/N=+\frac{1}{2}$ vs $N\tau$ for $N=20$. The solid line gives the exact calculation, the dashed line corresponds to the Gaussian field hypothesis, and the dotted line is obtained by using a classical radiation rate.

which is independent of N for large N, and

$$\left(\frac{d}{d\tau}\,g^{(2)}_{\rm inc}(\tau,0)\right)_{\tau=0}=0. \tag{18}$$

To have an idea of the variation of $g^{(2)}(\tau,0)$ against τ, we can take the value of $\langle R_+R_-\rangle$ given by the classical model[9]

$$\langle R_+R_-\rangle=\frac{N^3}{4(N-1)}\,\operatorname{sech}^2\frac{(N\tau-\tau_s)}{2}\,, \tag{19}$$

where $\tau_s=\ln(N-1)$. So, Eq. (13) becomes

$$g^{(2)}(\tau,0)\approx 1-\tanh\tfrac{1}{2}(N\tau-\tau_s)\,, \tag{20}$$

which gives a good enough idea of the correlations (see the dotted line in Fig. 3).

For the time τ_0, in order that $W(\tau_0)=0$, $g^{(2)}(\tau,\tau_0)$ decays from its initial value, roughly 1, to 0 when $N\tau$ increases (Fig. 4). The radiation field is not coherent. Knowing that the variance σ of the system's energy is maximum and approximately

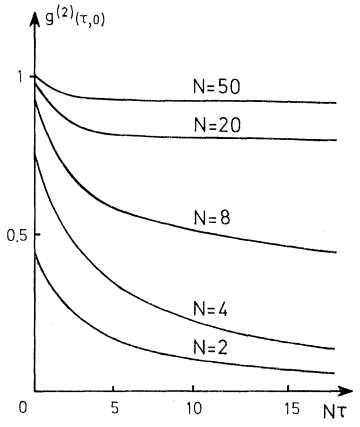

FIG. 5 Normalized intensity-fluctuation function $g^{(2)}(\tau,0)$ vs $N\tau$ for several systems prepared by intense laser $\frac{1}{2}\pi$ pulse.

equal to $N^2/16$ for zero energy,[10] the radiation rate

$$\langle R_+R_-\rangle = \tfrac{1}{2}N(\tfrac{1}{2}N+1) - \sigma + W - W^2 \approx \tfrac{3}{16}N^2 \qquad (21)$$

is proportional to N^2. So, at time τ_0, the state of our system obeys the superradiant conditions ($W \approx 0, \langle R_+R_-\rangle \approx N^2$). But these conditions do not seem to ensure a coherent radiation field.

We have also performed calculations with systems prepared by an intense laser $\frac{1}{2}\pi$ pulse, i.e.,

$$\rho_{m,m}(0) = \frac{N!}{(\frac{1}{2}N+m)!\,(\frac{1}{2}N-m)!}\,(2)^{-N}\,. \qquad (22)$$

The variation of $g^{(2)}(\tau, 0)$ against time $N\tau$ is plotted in Fig. 5 for different N. For sufficiently large N, $g^{(2)}(\tau, 0)$ decays from roughly unity to a nonzero value. A straightforward calculation shows that with the initial conditions given by Eq. (22), the relation (13), which expresses the correlations as a function of the radiation rates, is valid for large N[i.e., the quantity $\langle R_+(t_2)R_-(t_2)R_+(t_1)R_-(t_1)$ factorizes]. Then, by introducing the classical radiation rates[9] [Eq. (13)] the asymptotic value of $g^{(2)}(\tau, 0)$ is found to be $1-4/(N+1)$. Clearly, for a large system, $g^{(2)}(\tau, 0)$ is constant and equal to 1. The radiation field emitted by a system prepared as Eq. (22) is coherent (at least) to the second-order.

As the initial values of the energy and the radiation rate are approximately the same in the two cases illustrated by Figs. 4 and 5, the different behaviors of the intensity-fluctuation function are due to the $\rho_{m,m}(\tau_0)$ distribution.[2] If one hypothesizes that a property of a superradiant state is coherent emission. This implies not only conditions for the energy and radiation rate of the system but also for its preparation.

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Absence of Nonlinear Response in the Quasistochastic Model*

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The full nonlinear governing equations for the quasistochastic model, which describes the motion of a Brownian charge carrier in an external electric field, are solved for the stationary current response. We find that the nonlinear response is identical to the linear response obtained earlier by Lebowitz and Rubin.

I. INTRODUCTION

The classical theory of Brownian motion, which is based on an *ab initio* stochastic description, can be formulated in terms of the Fokker-Planck equation[1]

$$\frac{\partial f}{\partial t} + \vec{\mathbf{V}}\cdot\frac{\partial f}{\partial \vec{\mathbf{R}}} + \frac{e}{M}\,\vec{\mathbf{E}}\cdot\frac{\partial f}{\partial \vec{\mathbf{V}}} = D\frac{\partial}{\partial \vec{\mathbf{V}}}\cdot\left(f\,\frac{\partial}{\partial \vec{\mathbf{V}}}\ln f/f_0\right). \qquad (1)$$

In order to describe the higher-order response it is necessary to consider a more detailed description of Brownian motion than that given by (1). This is of interest since at present only formal results[2] for nonlinear response coefficients have been obtained in the literature. The case of conduction by Brownian charge carriers offers a physically relevant problem, e.g., for the area of electrolyte theory,[3] which should be easier to treat than the general problem of conduction by arbitrary carriers.

A generalization of (1) which eliminates the stochastic content in the description and proceeds instead directly from the Liouville equation has been obtained by several authors[4] (the *first* of these references will be referred to as LR hereafter). They find a kinetic equation for the Brownian par-