

Photon statistics in multiphoton absorption and emission processes

M. S. Zubairy

Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

J. J. Yeh

Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

(Received 26 November 1979)

Exact solutions of the master equations which describe single-mode m -photon absorption and m -photon emission processes in a two-level atomic system are obtained using a density-matrix approach. Some aspects of the photon statistics, e.g., antibunching, are discussed.

I. INTRODUCTION

In recent years, the study of the photon statistics in multiphoton processes has received a great deal of interest.¹⁻⁹ Several authors have discussed the possibility of observing antibunching in multiphoton absorption process.^{7,9}

Up to now exact expressions have only been derived for the photon distribution function in a two-photon absorption process using a generating function method and in a two-photon emission process using a Laplace transform method. The generating function approach is rather inconvenient in the study of photon statistics of m -photon processes when $m > 2$.

In this paper, we present exact solutions of the master equations which describe m -photon absorption and emission processes, using a density-matrix method. The diagonal elements of the density matrix determine the photon distribution function. By taking the appropriate moments with respect to the distribution function in the m -photon absorption process, we evaluate the photon number fluctuations. With the help of the figures we demonstrate that, for initial coherent and chaotic fields, the fluctuations decrease below unity when $m \geq 2$. This is a manifestation of photon antibunching.

II. MASTER EQUATION FOR THE DENSITY MATRIX OF THE FIELD

Let us consider a coupled system of field and N noninteracting two-level atoms. We assume that the atoms make transitions from lower (upper) level to the upper (lower) level by absorbing (emitting) m photons, one photon in each of the modes k_1, k_2, \dots, k_m . The number of atoms in the two levels are assumed to be maintained constant by some external influence. The interaction Hamil-

tonian for this process is expressed in the form¹

$$H_I = \sum_{i=1}^N \{ \eta_m \hat{C}_{2i}^\dagger \hat{C}_{1i} \hat{E}_{k_1}^\dagger(\vec{r}_i) \dots \hat{E}_{k_m}^\dagger(\vec{r}_i) + \text{H.c.} \}, \quad (1)$$

where \hat{C}_{1i} , \hat{C}_{2i} , \hat{C}_{1i}^\dagger , and \hat{C}_{2i}^\dagger are the annihilation and creation operators for the i th atom in the ground state $|1_i\rangle$ and the excited state $|2_i\rangle$. η_m is the matrix element for the m -photon transition. The positive-frequency part of the electric field at the i th atom is given by

$$\hat{E}_{k_i}^\dagger(\vec{r}_j) = -i(2\pi\hbar\omega_{k_i})^{1/2} U_{k_i}(\vec{r}_j) \hat{a}_{k_i}, \quad (2)$$

where ω_{k_i} is the frequency of the k_i th mode of the field (with atomic transition frequency $\omega = \omega_{k_1} + \dots + \omega_{k_m}$), $U_{k_i}(\vec{r}_j)$ is the mode function of the k_i th mode at the position \vec{r}_j , and \hat{a}_{k_i} is the photon destruction operator of the k_i th mode.

In the interaction picture, the equation of motion for the density operator $\hat{\rho}$ of the atom-field system is given by

$$i\hbar \frac{\partial}{\partial t} \hat{\rho} = [\hat{H}_I, \hat{\rho}]. \quad (3)$$

We assume that at some initial time t_0 , the radiation field and the atomic system are decoupled; i.e.,

$$\hat{\rho}(t_0) = \hat{\rho}_F(t_0) \otimes \prod_{i=1}^N \hat{\rho}_i(t_0), \quad (4)$$

where $\hat{\rho}_F(t_0) = \text{Tr}_A \hat{\rho}(t_0)$ is the reduced density matrix of the field and $\hat{\rho}_i(t_0)$ is the initial density operator of the i th atom at time t_0 .

The equation of motion for $\hat{\rho}_F(t)$ may be obtained using the standard perturbation techniques based on Born and Markoff approximations. The resulting equation is⁴

$$\begin{aligned} \frac{d\hat{\rho}_F}{dt} = & -\beta^{(m)}K_1 \left[\left(\prod_{i=1}^m \hat{a}_{k_i}^\dagger \right) \left(\prod_{j=1}^m \hat{a}_{k_j} \right) \hat{\rho}_F - 2 \left(\prod_{i=1}^m \hat{a}_{k_i} \right) \hat{\rho}_F \left(\prod_{j=1}^m \hat{a}_{k_j}^\dagger \right) + \hat{\rho}_F \left(\prod_{i=1}^m \hat{a}_{k_i}^\dagger \right) \left(\prod_{j=1}^m \hat{a}_{k_j} \right) \right] \\ & -\beta^{(m)}K_2 \left[\left(\prod_{i=1}^m \hat{a}_{k_i} \right) \left(\prod_{j=1}^m \hat{a}_{k_j}^\dagger \right) \hat{\rho}_F - 2 \left(\prod_{i=1}^m \hat{a}_{k_i}^\dagger \right) \hat{\rho}_F \left(\prod_{j=1}^m \hat{a}_{k_j} \right) + \hat{\rho}_F \left(\prod_{i=1}^m \hat{a}_{k_i} \right) \left(\prod_{j=1}^m \hat{a}_{k_j}^\dagger \right) \right], \end{aligned} \quad (5)$$

where K_1 and K_2 are the thermal populations of the atomic states $|1\rangle$ and $|2\rangle$; $\beta^{(m)}$ is given by

$$\begin{aligned} \beta^{(m)} = & (2\pi\hbar)^{m-2} (2\pi^2) \left(\prod_{i=1}^m \omega_{k_i} \right) |\eta_m|^2 g(\omega) \\ & \times \int d\vec{r} N(\vec{r}) \left(\prod_{i=1}^m |U_{k_i}(\vec{r})|^2 \right). \end{aligned} \quad (6)$$

In Eq. (6) $g(\omega)$ is the line-shape function of the atoms and $N(\vec{r})$ is the atomic density in the medium.

Although the method for the solution of the reduced density matrix of the field in m -photon absorption and emission processes in the following sections can be applied for arbitrary numbers of field modes, we shall restrict ourselves to a single-mode case (with $k_1 = k_2 = \dots = k_m$) for the sake of mathematical simplicity. Then Eq. (5) becomes

$$\begin{aligned} \frac{d\hat{\rho}_F}{dt} = & -\beta^{(m)}K_1 \{ \hat{a}^{\dagger m} \hat{a}^m \hat{\rho}_F - 2 \hat{a}^m \hat{\rho}_F \hat{a}^{\dagger m} + \hat{\rho}_F \hat{a}^{\dagger m} \hat{a}^m \} \\ & -\beta^{(m)}K_2 \{ \hat{a}^m \hat{a}^{\dagger m} \hat{\rho}_F - 2 \hat{a}^{\dagger m} \hat{\rho}_F \hat{a}^m + \hat{\rho}_F \hat{a}^m \hat{a}^{\dagger m} \}. \end{aligned} \quad (7)$$

It is worthwhile to mention that this equation describes an m -photon laser¹⁰ in linear approximation when the cavity losses are simulated by m -photon absorption process to achieve detailed balance. We shall, however, restrict ourselves in the next sections to m -photon absorption case ($k_1 = 1, k_2 = 0$) and m -photon emission case ($k_1 = 0, k_2 = 1$) separately.

III. m -PHOTON ABSORPTION

If we calculate the matrix element of each term in Eq. (7) between Fock states $\langle n|$ and $|n+k\rangle$

$$\begin{aligned} \partial \rho(nm+i, nm+k+i, \tau) / \partial \tau = & a(nm+i, nm+k+i) \rho(nm+i, nm+k+i, \tau) \\ & + b(nm+m+i, nm+m+k+i) \rho(nm+m+i, nm+m+k+i, \tau), \end{aligned} \quad (11)$$

with $i = 0, 1, \dots, m-1$. In matrix notation these equations are written as

$$\frac{\partial}{\partial \tau} \rho_i(\tau) = M_i \rho_i(\tau), \quad (12)$$

where

($k = 0, 1, 2, \dots$), the matrix element $\rho(n, n+k, \tau) = \langle n | \rho_F(\tau) | n+k \rangle$ of the reduced density matrix ρ_F satisfies an equation of motion of the form (for $K_1 = 1, K_2 = 0$):

$$\begin{aligned} \partial \rho(n, n+k, \tau) / \partial \tau = & a(n, n+k) \rho(n, n+k, \tau) \\ & + b(n+m, n+k+m) \\ & \times \rho(n+m, n+k+m, \tau), \end{aligned} \quad (8)$$

where $\tau = 2\beta^{(m)}t$ and

$$a(n, n+k) = -\frac{1}{2} \left(\frac{n!}{(n-m)!} + \frac{(n+k)!}{(n+k-m)!} \right), \quad (9a)$$

$$b(n, n+k) = \left(\frac{n!(n+k)!}{(n-m)!(n-m+k)!} \right)^{1/2}. \quad (9b)$$

The matrix element $\rho(n+k, n, \tau)$ is just the complex conjugate of $\rho(n, n+k, \tau)$; i.e.,

$$\rho(n+k, n, \tau) = \rho^*(n, n+k, \tau). \quad (10)$$

The solution of Eq. (8) would, therefore, completely determine the reduced density matrix $\hat{\rho}_F$ of the field.

It is apparent from Eq. (8) that an element $\rho(n+i, k+n+i, \tau)$ ($i = 0, 1, \dots, m-1$) is related only to the elements $\rho(n+i, k+n+i, \tau)$, $\rho(n+i+m, k+n+i+m, \tau)$, and $\rho(n+i+2m, k+n+i+2m, \tau) \dots$ as shown in Fig. 1. We can therefore replace Eq. (8) by the following set of m independent equations corresponding to m chains in Fig. 1:

$$\rho_i(\tau) = \begin{pmatrix} \rho(i, k+i, \tau) \\ \rho(m+i, m+k+i, \tau) \\ \vdots \\ \rho(nm+i, nm+k+i, \tau) \\ \vdots \\ \vdots \end{pmatrix}, \tag{13}$$

$$M_i = \begin{pmatrix} a(i, k+i) & b(m+i, m+k+i) & & 0 \\ & a(m+i, m+k+i) & b(2m+i, 2m+k+i) & \\ & & \vdots & \vdots \\ & & & \vdots \\ 0 & & & \end{pmatrix}. \tag{14}$$

We now define the eigenvalue $\lambda_{l,i}$ of the matrix corresponding to the right eigenstate

$$A_{l,i} = \begin{pmatrix} \alpha_0^{l,i} \\ \alpha_1^{l,i} \\ \vdots \\ \vdots \\ \vdots \end{pmatrix} \tag{15}$$

and the left eigenstate

$$B_{l,i} = (\beta_0^{l,i}, \beta_1^{l,i}, \dots), \tag{16}$$

by the following relations:

$$M_i A_{l,i} = \lambda_{l,i} A_{l,i}, \tag{17}$$

$$B_{l,i} M_i = \lambda_{l,i} B_{l,i}. \tag{18}$$

If we multiply Eq. (17) by $B_{l',i}$ from the left and Eq. (18) for l' by $A_{l,i}$ from the right and subtract the two equations, we get an orthogonality relation:

$$B_{l',i} A_{l,i} = \delta_{ll'}, \tag{19}$$

when $\lambda_{l,i} \neq \lambda_{l',i}$.

In view of Eq. (17), the solution of Eq. (12) is given by

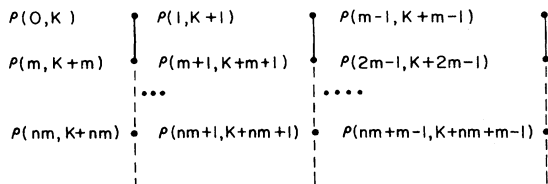


FIG. 1. The chain structure of Eq. (8).

$$\rho_i(\tau) = \sum_{l=0}^{\infty} C_l A_{l,i} e^{\lambda_{l,i} \tau}. \tag{20}$$

In case the eigenvalues are nondegenerate, the coefficient C_l can be determined using the orthogonality condition (16), and we obtain

$$C_l = B_{l,i} \rho_i(0). \tag{21}$$

Equations (20) and (21) can be recast in the functional form by the following equation:

$$\rho(mn+i, mn+k+i, \tau) = \sum_{l=0}^{\infty} \sum_{q=0}^{\infty} \beta_q^{l,i} \alpha_n^{l,i} e^{\lambda_{l,i} \tau} \rho(mq+i, mq+k+i, 0). \tag{22}$$

It is therefore evident that a determination of the eigenvalues $\lambda_{l,i}$ and the right and left eigenstates $A_{l,i}$, $B_{l,i}$ of the matrix M_i would completely determine the reduced density matrix $\hat{\rho}_F$.

The eigenvalues $\lambda_{l,i}$ of M_i , which satisfy the equation

$$\det(M_i - \lambda I) = 0 \tag{23}$$

(I being the unit matrix), are given by

$$\lambda_{l,i} = a(ml+i, ml+k+i) = -\frac{1}{2} \left(\frac{(ml+i)!}{(ml+i-m)!} + \frac{(ml+k+i)!}{(ml+k+i-m)!} \right). \tag{24}$$

It is easy to verify that $\lambda_{l,i} \neq \lambda_{l',i}$ for $l \neq l'$. On substituting from Eq. (24) into Eq. (17), we get an infinite set of equations whose n th member is the following recursion relation:

$$a(mn - m + i, mn - m + k + i)\alpha_{n-1}^{l,i} + b(mn + i, mn + k + i)\alpha_n^{l,i} = a(ml + i, ml + k + i)\alpha_{n-1}^{l,i}. \tag{25}$$

By iterating this recursion relation, we obtain

$$\alpha_n^{l,i} = \begin{cases} \alpha_0^{l,i} \prod_{r=1}^n \frac{[a(ml+i, ml+k+i) - a(mr-m+i, mr-m+k+i)]}{b(mr+i, mr+k+i)}, & n \leq l \\ 0, & n > l. \end{cases} \tag{26}$$

In a similar manner, it follows from Eqs. (18) and (24) that

$$\beta_q^{l,i} = \begin{cases} \beta_l^{l,i} \prod_{r=l+1}^q \frac{b(mr+i, mr+k+i)}{[a(ml+i, ml+k+i) - a(mr+i, mr+k+i)]}, & q \geq l \\ 0, & q < l. \end{cases} \tag{27}$$

To determine $\alpha_0^{l,i}$ and $\beta_l^{l,i}$ we first notice that, in view of Eqs. (15), (16), and (19), the following relationship holds:

$$B_{l,i} A_{l,i} = \beta_l^{l,i} \alpha_l^{l,i} = 1. \tag{28}$$

We can therefore choose

$$\alpha_l^{l,i} = 1, \tag{29a}$$

$$\beta_l^{l,i} = 1. \tag{29b}$$

It then follows from Eqs. (26) and (29a) that

$$\alpha_0^{l,i} = \prod_{r=1}^l \frac{b(mr+i, mr+k+i)}{[a(ml+i, ml+k+i) - a(mr-m+i, mr-m+k+i)]}. \tag{30}$$

The choices (29b) and (30) enable us to combine the m equation in Eq. (22) corresponding to m values of i (i.e., $i = 0, 1, \dots, m - 1$).

On substituting from Eqs. (24), (26), (27), (29b), and (30) into Eq. (22), we obtain, after some rearrangement,

$$\rho(n, n', \tau) = \sum_{l=n}^q \sum_{q=l}^{\infty} \frac{\left(\prod_{r=n+m}^q b(r, r+n'-n) \right) e^{a(l, l+n'-n)\tau} \rho(q, q+n'-n, 0)}{\left(\prod_{r=n}^q [a(l, l+n'-n) - a(r, r+n'-n)] \right)}, \tag{31}$$

where the primes on the summation and product signs mean that the increment is in steps of m . This equation combined with the expressions of $a(r, r')$ and $b(r, r')$, i.e.,

$$a(r, r') = -\frac{1}{2} \left(\frac{r!}{(r-m)!} + \frac{r'!}{(r'-m)!} \right), \tag{32}$$

$$b(r, r') = \left(\frac{r! r'!}{(r-m)! (r'-m)!} \right)^{1/2}, \tag{33}$$

completely determines the evolution of the reduced density matrix of the field in the m -photon absorption process.

The photon distribution function $p(n, \tau)$, which is obtained by putting $n = n'$ in Eq. (31), is given by

$$p(n, \tau) = \sum_{l=n}^{\infty} \sum_{q=l}^{\infty} \frac{\left(\prod_{r=n+m}^q b(r, r) \right) e^{a(l, l)\tau} p(q, 0)}{\left(\prod_{r=n, r \neq l}^q [a(l, l) - a(r, r)] \right)}. \tag{34}$$

It is not difficult to verify that, for the two-photon absorption process (i.e., for $m = 2$), this expression for $p(n, \tau)$ reduces identically to the one obtained

by Simann and Loudon⁷ using the generating function method. In Figs. 2-7, we have plotted $p(n, \tau)$ versus n for various values of τ in two-, three-,

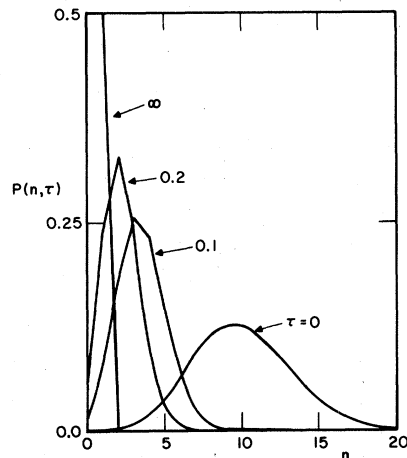


FIG. 2. $p(n, \tau)$ versus n for two-photon absorption process with initial coherent state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.1, 0.2$, and ∞ , respectively.

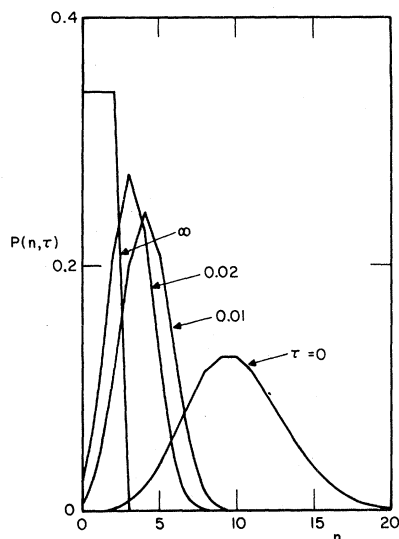


FIG. 3. $p(n, \tau)$ versus n for three-photon absorption process with initial coherent state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.01, 0.02$, and ∞ , respectively.

and four-photon processes when the initial photon distributions are coherent and chaotic, i.e.,

$$p_{\text{coh}}(n, 0) = \frac{\bar{n}_0^n e^{-\bar{n}_0}}{n!}, \quad (35)$$

$$p_{\text{cha}}(n, 0) = \frac{\bar{n}_0^n}{(1 + \bar{n}_0)^{1+n}}. \quad (36)$$

With the expression (34) for $p(n, \tau)$, the expectation values $\langle n^r \rangle$ ($r = 1, 2, \dots$) for the photon numbers can be evaluated using the formula

$$\langle n^r \rangle = \sum_{n=0}^{\infty} n^r p(n, 0). \quad (37)$$

In Figs. 8 and 9, we have plotted the quantity

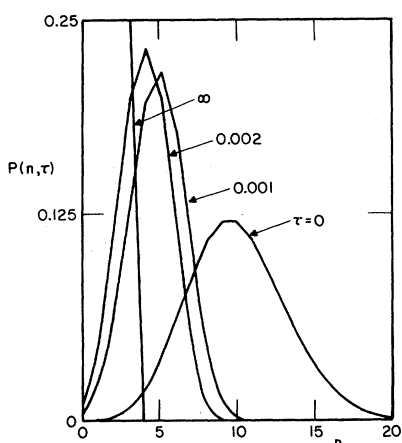


FIG. 4. $p(n, \tau)$ versus n for four-photon absorption process with initial coherent state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.001, 0.002$, and ∞ , respectively.

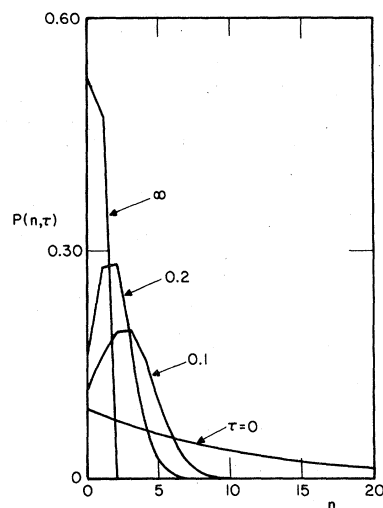


FIG. 5. $p(n, \tau)$ versus n for two-photon absorption process with initial chaotic state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.1, 0.2$, and ∞ , respectively.

$$g_2(\tau) = \frac{\langle n^2 \rangle - \langle n \rangle^2}{\langle n \rangle} \quad (38)$$

versus τ in two-, three-, and four-photon absorption processes for initial coherent and chaotic distributions. (Caution must be used in interpreting the results from these figures because $\tau = 2\beta^{(m)}t$ depends upon m .)

It is evident from the figures that, in many cases, $g_2(\tau) < 1$. This is a manifestation of the photon antibunching. Simaan and Loudon⁷ discussed this effect in two-photon absorption process and Paul, Mohr, and Brunner⁹ predicted this effect in an arbitrary m -photon absorption process (with $m > 2$) using a perturbative method.

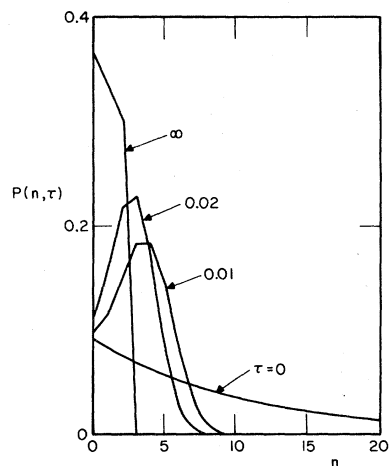


FIG. 6. $p(n, \tau)$ versus n for three-photon absorption process with initial chaotic state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.01, 0.02$, and ∞ , respectively.

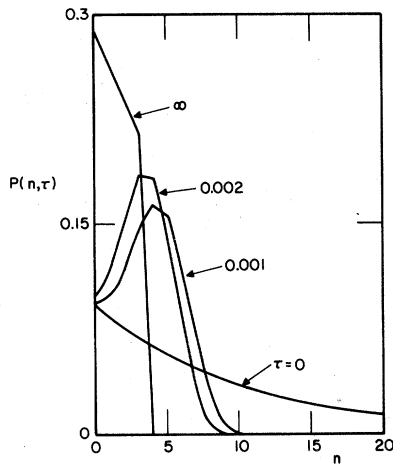


FIG. 7. $p(n, \tau)$ versus n for four-photon absorption process with initial chaotic state (with $\bar{n}_0 = 10$) at $\tau = 0, 0.001, 0.002$, and ∞ , respectively.

It follows from Eq. (34) that, in the steady state ($\tau \rightarrow \infty$), we obtain

$$p(n, \infty) = \begin{cases} 0, & n \geq m \\ \sum_{q=n}^{\infty} \prod_{r=n+q}^q \frac{b(r, r)}{[a(n, n) - d(r, r)]}, & n < m. \end{cases} \quad (39)$$

This expression for $p(n, \infty)$ is according to our expectation that the steady state photon distribution function is nonvanishing only for $n < m$. In Table I we have presented the values of $g_2(\infty)$ and $\langle n \rangle_{\tau=\infty}$ in two-, three-, and four-photon absorption processes with initial coherent and chaotic photon distribution functions.

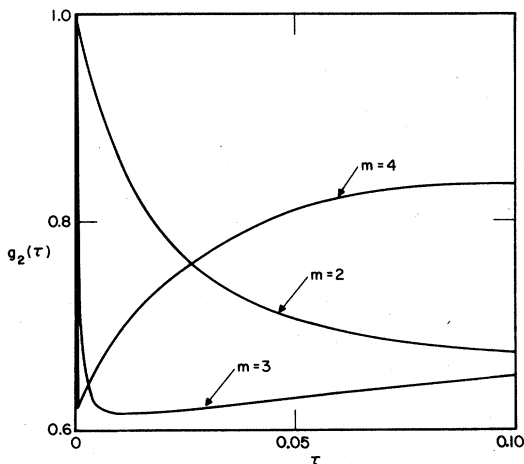


FIG. 8. $g_2(\tau)$ versus τ for two-, three-, and four-photon absorption processes with initial coherent state (with $\bar{n}_0 = 10$).

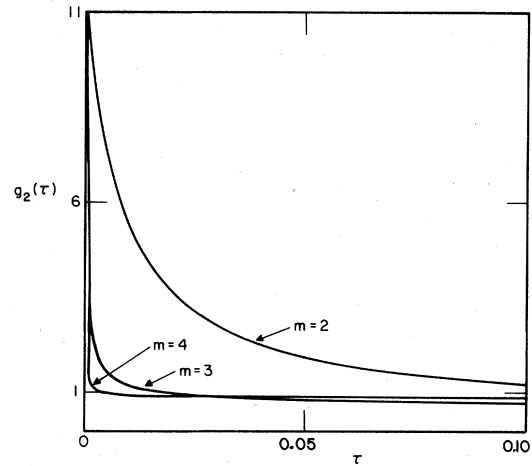


FIG. 9. $g_2(\tau)$ versus τ for two-, three-, and four-photon absorption processes with initial chaotic state (with $\bar{n}_0 = 10$).

IV. m -PHOTON EMISSION

We now consider the equation of motion of the matrix element $\rho(n, n+k, \tau)$ of the reduced density matrix $\hat{\rho}_F$ in the case of the m -photon emission process which is obtained from Eq. (7) with $K_1 = 0$ and $K_2 = 1$. It is given by

$$\begin{aligned} \partial \rho(n, n+k, \tau) / \partial \tau = & c(n, n+k) \rho(n, n+k, \tau) \\ & + d(n-m, n+k-m) \\ & \times \rho(n-m, n+k-m), \end{aligned} \quad (40)$$

where

$$c(n, n+k) = -\frac{1}{2} \left(\frac{(n+m)!}{n!} + \frac{(n+k+m)!}{(n+k)!} \right), \quad (41)$$

$$d(n, n+k) = \left(\frac{n!(n+k)!}{(n-m)!(n+k-m)!} \right)^{1/2}. \quad (42)$$

Following the method outlined in the previous section, we first replace Eq. (40) by the following set of m independent equations, for $i=0, 1, 2, \dots, m-1$:

TABLE I. The value of $g_2(\infty)$ and $\langle n \rangle_{\tau=\infty}$ for several multiphoton absorption processes with coherent and chaotic initial photon distributions.

	Coherent $\bar{n}_0 = 10$		Chaotic $\bar{n}_0 = 10$	
	$g_2(\infty)$	$\langle n \rangle_{\tau=\infty}$	$g_2(\infty)$	$\langle n \rangle_{\tau=\infty}$
Two-photon process	0.500	0.5	0.524	0.476
Three-photon process	0.667	1.0	0.709	0.936
Four-photon process	0.833	1.5	0.898	1.381

- ¹Y. R. Shen, Phys. Rev. 155, 921 (1967).
²P. Lambropoulos, Phys. Rev. 156, 286 (1967).
³B. R. Mollow, Phys. Rev. 175, 1555 (1968).
⁴G. S. Agarwal, Phys. Rev. A 1, 1445 (1970).
⁵K. J. McNeil and D. F. Walls, J. Phys. A 7, 617 (1974).
⁶N. Tornaou and A. Bach, Opt. Commun. 11, 46 (1974).
⁷H. D. Simaan and R. Loudon, J. Phys. A 8, 539 (1975);
8, 1140 (1975).
⁸A. Bandilla and H. H. Ritze, Phys. Lett. 55A, 285 (1975).
⁹H. Paul, U. Mohr, and W. Brunner, Opt. Commun. 17, 145 (1976).
¹⁰K. J. McNeil and D. F. Walls, J. Phys. A 8, 104 (1975).