

## Quantum statistics of a multiphoton-laser amplifier at low intensity level

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Numerical results for the statistical properties, and time-intensity correlation function, of a multiphoton-laser amplifier operating at very low intensity level are obtained and analyzed. Comparison with the rotating van der Pol oscillator model is made. Time variation of the second-order cumulant of the photon number displays a maximum at a time which is approximately evaluated.

### I. INTRODUCTION

Photon statistics of a single-mode laser radiation continue to be the subject of several publications both experimental and theoretical. As a matter of fact, in seeking the most precise description of the laser field, it is necessary to study all the physical processes involved in the interaction between atoms and field. For instance, effect of cooperative atomic interactions on photon statistics has been examined by Huang and Mandel<sup>1</sup> and subsequently by Zubairy.<sup>2</sup> More recently contributions from multiphoton processes have been investigated but exact results are well established only when absorption and emission processes are studied separately.<sup>3</sup> If both processes are taken into account, analytical solutions for the master equation which describes the evolution of the diagonal elements of matrix density of the field are difficult to obtain except for the steady-state solution and within the condition of detailed balance between absorption and emission.<sup>4</sup> Very recently, in order to investigate possible relations between photon antibunching and strong saturation, Mohr<sup>5</sup> has studied the photon statistics in saturated  $M$ -photon amplification and attenuation of coherent light. But here again effects of emission and absorption have been analyzed distinctly and, to obtain analytical results, only the case of high intensity level has been considered. It is the purpose of this paper to treat the general problem and to present numerical results on the photon statistics of multiphoton lasers which obey the detailed balance condition at the low intensity level.

### II. GENERAL EQUATIONS

We consider some interactions between a field and a homogeneous set of  $N$ -independent two-levels quantum systems. For the sake of simplicity we shall call such a system an atom. We assume that the atoms make transitions between these levels, and  $M$ -photon processes of absorption and emission are possible. To simplify the theory we restrict ourselves to the case of a single-mode field whose frequency is resonant with the atom transition frequency. We suppose that the number of excited atoms can be held constant by some external pumping process.

#### A. Master equation

Following the method initiated by Scully and Lamb<sup>6</sup> for the one-photon laser, it is possible to derive the equation of motion for  $\rho_f$  the density matrix of the field and then to obtain the master equation governing the evolution of the diagonal elements  $P(n,t) = \langle n | \rho_f(t) | n \rangle$ , which is the probability distribution to find  $n$  photons in the field at time  $t$ . Let us very briefly outline the method.

In the interaction picture, we start with

$$i\hbar \frac{\partial}{\partial t} \rho_{af}(t) = [H, \rho_{af}(t)], \quad (1)$$

where  $H$  is the interaction Hamiltonian and  $\rho_{af}(t)$  is the density operator of the atom-field system. It is usual to assume that the field and atoms are uncorrelated at the initial time  $t_0$ . Scully and Lamb have shown, using perturbation methods, that Eq. (1) can be rewritten as

$$\rho_f^{(r)}(t_0+t) = \rho_f(t_0) + \text{Tr}_a \left[ \sum_{p=1}^r \left[ \frac{-i}{\hbar} \right]^p \int_{t_0}^t dt_1 \cdots \int_{t_0}^{t_{p-1}} dt_p [H(t_1), [H(t_2), \dots [H(t_p), \rho_{af}(t_0)] \cdots ] ] \right] \quad (2)$$

with  $\rho_f(t_0) = \text{Tr}_a \{ \rho_{af}(t_0) \}$ . Now, Eq. (2) requires solution of the expression of  $H$ . Under the rotating-wave approximation and for the case of interest here, we make use of

$$H = \hbar g_M \begin{pmatrix} 0 & a^M \\ (a^\dagger)^M & 0 \end{pmatrix}, \quad (3)$$

where  $g_M$  is the coupling constant for an  $M$ -photon dipole transition,  $a$  is the photon annihilation operator, and  $a^\dagger$  is the photon creation operator. With the given initial distribution  $\rho_{af}(t_0)$  for the atom-field system and the decorrelation condition

$$\rho_{af}(t_0) = \rho_f(t_0) \otimes \rho_a(t_0) = \begin{pmatrix} N_a \rho_f(t_0) & 0 \\ 0 & N_b \rho_f(t_0) \end{pmatrix}, \quad (4)$$

where  $N_a$  and  $N_b$  are constants. Therefore the calculation of (1) using (2) is done with the help of the property (55) of the Ref. 6 (see p. 295). For  $r=2$ , in Eq. (2) with (3) and (4), the equation obtained describes an  $M$ -photon laser in linear approximation. As we expand Eq. (2) to the fourth-order approximation ( $r=4$ ), terms of saturation begin to be non-negligible. For higher-order terms in Eq. (2), the master equation for the matrix elements  $P_M(n, t)$  of  $\rho_f$  could be written in the form given by McNeil and Walls.<sup>4</sup> It is an appropriate generalization of the  $M=1$  equation [see Eq. (17), p. 285 of Ref. 6]

$$\begin{aligned} \frac{dP_M(n, t)}{dt} = & - \prod_{l=1}^M (n+l) [F_M(n) P_M(n, t) - \beta_M P_M(n+M, t)] \\ & + \prod_{l=0}^{M-1} (n-l) [G_M(n) P_M(n-M, t) - \beta_M P_M(n, t)], \end{aligned} \quad (5)$$

where

$$F_M(n) = \left[ 1 + \chi_M \prod_{l=1}^M (n+l) \right]^{-1}, \quad (6)$$

and

$$G_M(n) = \left[ 1 + \chi_M \prod_{l=1}^{M-1} (n-l) \right]^{-1}; \quad (7)$$

$\beta_M$  is the loss parameter and  $\chi_M$  is the saturation parameter which is essentially proportional to  $g_M^2$ . The time  $t$  is defined by  $t = A_M z$  where  $A_M$  is the gain parameter. Very recently Eq. (5) has been established in a different way by Singh (see Ref. 7).

The general solution of Eq. (5) is difficult to obtain except in some special situations. For example, if only absorption (or emission) terms are considered, exact results are reached and given in (Ref. 3). Also by invoking detailed balance considerations and given the initial photon distribution depending only on  $P(nM)$ ,  $n=0, 1, \dots$ , the steady-state solution  $P_s(n)$  can then be evaluated. One obtains<sup>4</sup>

$$P_s(nM) = P_s(0) \prod_{k=1}^n \frac{1}{\beta_M \left[ 1 + \chi_M \prod_{j=0}^{M-1} (kM-j) \right]}, \quad (8a)$$

$$P_s(nM+j) = 0; \quad (8b)$$

for  $n=0, 1, 2, \dots$  and  $j=1, 2, \dots, M-1$ .

### B. Intensity correlation function

Since it is interesting to understand how the photon statistics are modified by the  $M$ -photon processes, it is also worthwhile to study how the multiphoton processes affect the time-dependent properties of the field.

It is usual to associate with Eq. (5) the continuous nonstationary Fokker-Planck-type equation. A rigorous derivation of this equation is not simple. However, it is a useful way to determine the evolution of the laser not too far from the threshold. Therefore given the  $P_0(I, t_0)$  initial intensity probability distribution, the  $P(I, t)$  describes completely the dynamical evolution of the light intensity. It is also usual to seek  $P(I, t)$  in the product form

$$P(I, t) = P(I) T(t), \quad (9)$$

so that the problem consists in solving the eigenvalue equation

$$L P_q(I) = -\lambda_q P_q(I), \quad (10)$$

where  $L$  is the appropriate operator and  $\lambda_q$  the

eigenvalues. For the classical model of rotating-wave van der Pol oscillator (RWvP),<sup>8-10</sup>

$$L = -\frac{\partial}{\partial I}(2I^2 - 2aI - 4) + \frac{\partial^2}{\partial I^2}(4I), \quad (11)$$

where  $a$  is the pump parameter.

With the  $\delta$  function as the initial distribution,  $P(I, t)$  is then the transition probability  $P(I, t/I_0, t_0)$  given in the form<sup>11,12</sup>

$$P(I, t/I_0, t_0) = \sum_{q=0}^{\infty} \frac{P_q(I)P_q(I_0)}{P_s(I_0)} \times \exp[-\lambda_q(t-t_0)], \quad (12)$$

where  $P_s(I_0)$  is the stationary light intensity distribution. Now it is possible to calculate the required intensity time correlation function using the relations

$$P(I_2, t_2; I_1, t_1) = P(I_2, t_2/I_1, t_1)P(I_1, t_1) \quad (13)$$

and

$$\langle I_1 I_2 \rangle = \int \int I_1 I_2 P(I_2, t_2; I_1, t_1) dI_1 dI_2, \quad (14)$$

where

$$I_1 = I(t)$$

and

$$I_2 = I(t + \tau).$$

For the Markovian processes that are of interest here, it is well known that Eq. (14) actually takes the simpler form

$$G(\tau) = \langle I(t)I(t + \tau) \rangle = \sum_{q=0}^{\infty} C_q^2 e^{-\lambda_q |\tau|} \quad (15)$$

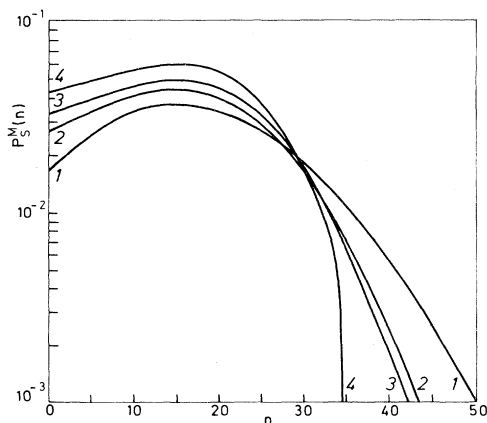


FIG. 1. Steady-state photon probability distribution  $P_s^M(n)$  for  $M=1, 2, 3$ , and 4.

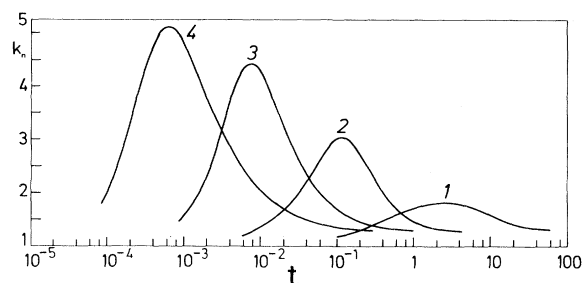


FIG. 2. Variation of the reduced second-order cumulant of the photon number vs undimensioned time for  $M=1, 2, 3$ , and 4.

with

$$C_q = \int_0^{\infty} I P_q(I, t) dI.$$

The behavior of the normalized correlation function given by Eq. (15),

$$\Lambda(\tau) = [G(\tau) - G(\infty)] / [G(0) - G(\infty)], \quad (16)$$

will be computed for various  $M$  at time  $t$  for which the stationary region is well approached.

### III. RESULTS AND DISCUSSION

Equation (5) has been solved numerically using the computational method described in Appendix A. It is adapted from the method that we already used for the one-photon process.<sup>13</sup>

The main results we want to discuss are summarized in Figs. 1–4. The values of the parameters are determined so that the mean number of photons at the steady state is approximately the same for all values of  $M$  and the loss parameter is chosen constant for all  $M$ :  $\beta=0.9$ ,  $\chi_1=7.1 \times 10^{-3}$ ,  $\chi_2=5 \times 10^{-4}$ ,  $\chi_3=2.9 \times 10^{-5}$ , and  $\chi_4=1.85 \times 10^{-6}$ . The mean number of photons at the origin of time axis  $\langle n \rangle_0$  is taken equal to 1.

In Fig. 1, we plot semilogarithmically the steady-

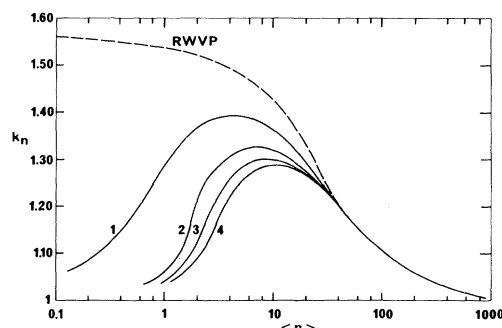


FIG. 3. Variation of the reduced second-order cumulant of the photon number calculated at the steady-state regime vs the mean number of photons for  $M=1, 2, 3$ , and 4.

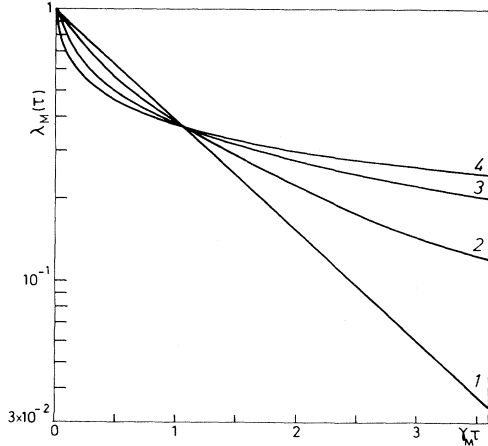


FIG. 4. Behavior of the reduced-time intensity correlation functions vs the reduced time  $\gamma_M \tau$  for  $M=1, 2, 3$ , and 4.

state solutions to Eq. (5) for  $M=1, \dots, 4$ . These have been obtained for  $t_1=60$ ,  $t_2=5$ ,  $t_3=0.9$ , and  $t_4=0.3$ . As recalled with Eqs. (5)–(7), the model chosen here for the  $M$ -photon amplifier is a process involving exchange of blocks of  $M$  photons. Therefore the curves are plotted for  $n=0, M, 2M, \dots, qM$ . The probability for other  $n$ 's is, according to (8), equal to 0. This is the case when the initial distribution is the Kronecker symbol. However, the numerical results show that  $P_s(qM+j)$  are very small but not equal to 0 for the initial distribution of Poisson (see Appendix A). All the results given here are obtained with the initial distribution of Poisson.

We point out that all distributions are not correctly fitted by a Gaussian shape. The Poisson distribution does not fit either the steady-state distribution for  $M=1$ . This deviation has already been noted for high  $\langle n \rangle_0$ . Also the limit value for the mean number of photons which has been taken as

$$\langle n \rangle_\infty = (\Delta/\beta\chi_M)^{1/M}, \quad (17)$$

where

$$\Delta = 1 - \beta \quad (18)$$

is not correctly verified for  $M \geq 2$ . So in order to compare our numerical results with the theory developed for high intensity level, we computed the normalized second cumulant of the photon number

$$k_n = \langle n(n-1) \rangle / \langle n \rangle^2. \quad (19)$$

It is well known that this quantity is identical to the normalized intensity variance

$$k_I = (\langle I^2 \rangle - \langle I \rangle^2) / \langle I \rangle^2. \quad (20)$$

The coefficient  $k_I$  is calculated from the RWvP

model as a function of the pump parameter  $a$  [see Eqs. (9) and (10) of Ref. 10]. In our notation

$$a = \Delta / \left[ \frac{\chi}{2} \right]^{1/2} \quad (21a)$$

and

$$P(I) = P_0 \exp \left[ \Delta I - \frac{\chi}{2} I^2 \right], \quad (21b)$$

where  $P_0$  is obtained from  $\int_0^\infty P(I) dI = 1$ .

In Fig. 2, the curves  $k_n$  for  $M=1, 2, 3$ , and 4 are plotted semilogarithmically versus  $t$ . We observe that for all  $M$ , the curves  $k_n(t)$  display a maximum at a time  $t_M$  which can be roughly approximated as shown in Appendix C. This approximation scheme seems reasonably good for  $M=1, 2$ . For other  $M$ , the discrepancy comes from the exponential behavior retained for the gain which is valid only for the very-short-time range. The exponential assumption is then clearly inadequate. Also it is important to note that for  $\langle n \rangle_0 > \langle n \rangle_\infty$  with  $\langle n \rangle_\infty$  given by (17) there is no maximum for  $k_n(t)$ , and  $k_n(t)$  increases smoothly from 1 to  $k_n(\infty) \simeq 1.30$ .

In order to test the validity of the RWvP model for  $M=1$ , we plot  $k_n$  vs  $\langle n \rangle$  for the steady-state regime in Fig. 3 together with the  $k_I$  derived from (20) and (21b). Now, for  $M=1, \dots, 4$ , we note that for  $\langle n \rangle \geq 40$ ,  $k_n$  given by our results and those derived from the RWvP model are in excellent agreement for all  $M$ . We notice first that  $k_n(\langle n \rangle)$  displays a maximum for  $5 < \langle n \rangle < 10$  and  $M$  varying from 1 to 4. Also at high  $\langle n \rangle$ ,  $k_n$  is independent of  $M$ , although  $\langle n \rangle$  and  $\langle n^2 \rangle$  are functions of  $M$ . For small values of  $\langle n \rangle$ , the deviation becomes larger and the RWvP model is clearly incorrect. Specially for  $\langle n \rangle \sim 0.1$ , the RWvP model leads to  $k_n \sim \pi/2$  instead of  $k_n \sim 1$ , which is the exact result that we have found numerically.

On the other hand, as recalled with the Eq. (15), the intensity correlation function, given  $M$ , can be expressed as a series of decreasing exponentials. The coefficients appearing in Eq. (15) can be calculated (Appendix B) in terms of the eigenfunctions  $P_q(I, t)$  given by Eq. (12). The behavior of the normalized-time intensity correlation functions given by (16) for each  $M$  is illustrated with the Fig. 4, where  $\gamma_M$  is the inverse of the time correlation. This has been calculated as the abscissa at which  $\Lambda_M(\tau_M) = e^{-1}$ . We found  $\tau_1 = 7.5$ ,  $\tau_2 = 0.14$ ,  $\tau_3 = 6 \times 10^{-2}$ , and  $\tau_4 = 5 \times 10^{-3}$ . Another definition of  $\tau_M$  [see Eq. (5-28) of Ref. 9] would not give very different results. We see that for  $M=1$ ,  $\lambda_1(\tau)$  is very close to a single exponential profile and as  $M$  increase the profile tends to be of constant value except in the immediate vicinity of the origin of the time axis. We found

that the eigenvalues  $\lambda_q$ , which are of importance in (15), are for  $q=1, \dots, M$  together with  $\lambda_0=0$ . Moreover, for  $M \rightarrow \infty$ ,  $\tau_M \sim 1/M$  so that

$$\Lambda_M(\tau) \sim c_0^2 + c_M^2 e^{-|\tau|/M} \quad (22)$$

as it begins to be verified for  $M=4$ .

In summary, we have considered and numerically studied the statistical properties of  $M$  photon laser amplifier at low intensity level and it is found that the RWvP model is not valid at very low intensity level. It is also proved that the correlation time decreases with  $M$  and shown that, for high  $M$ , each block of  $M$  photons can be treated independently.

#### APPENDIX A: COMPUTATIONAL METHOD FOR CALCULATION OF THE PROBABILITIES $P(n, t)$ DEFINED BY EQ. (5)

The system of  $N \times M$  first-order linear differential equations governing the time evolution of the distribution  $P(n, t)$  in a  $M$  photons amplifier, as defined by Eq. (5), can be written in the closed form [ $P(n, t) \equiv P_n(t)$ ],

$$\frac{dP^{(k)}(t)}{dt} = \hat{A}^{(k)} P^{(k)}(t), \quad k=0, \dots, M-1 \quad (A1)$$

where  $P^{(k)}$  is a vector of dimension  $N$  whose components are the functions  $P_{nM+k}(t)$ . The associated matrix  $\hat{A}^{(k)}$  has its elements defined by Eqs. (5)–(7). They do not depend on the time. Each system of equations defined by Eq. (A1) can be solved independently. The Laplace-transform method allows us to solve easily these equations. The Laplace transform of Eq. (A1) is

$$s\tilde{P}^{(k)}(s) - P^{(k)}(0) = \hat{A}^{(k)} P^{(k)}(s), \quad (A2)$$

where  $\tilde{P}^{(k)}(s)$  is a vector whose components are the Laplace transform of the components of  $P^{(k)}(t)$ ,  $s$  is the variable associated with  $t$  in the Laplace transform. The vector  $P^{(k)}(0)$  contains the initial value of  $P^{(k)}(t)$ .

The formal solution of Eq. (A2) can be written

$$\tilde{P}^{(k)}(s) = (sI - \hat{A}^{(k)})^{-1} P^{(k)}(0), \quad (A3)$$

where  $I$  is the  $N \times N$  identity matrix.

We assume that any matrix  $\hat{A}^{(k)}$  is nondefective. Therefore it has a set of  $N$  “right” (or “column”) eigenvectors  $\{|i, k\rangle\}$  associated with the  $N$  eigenvalues  $\lambda_i^{(k)}$  and defined by the identities

$$\hat{A}^{(k)} |i, k\rangle = \lambda_i^{(k)} |i, k\rangle. \quad (A4)$$

The space of vectors  $|i, k\rangle$  is complete and it has a

dual space of  $N$  “left” (or “row”) eigenvectors defined

$$\langle i, k | \hat{A}^{(k)} = \lambda_i^{(k)} \langle i, k |. \quad (A5)$$

We recall the usual relations between the vectors of the dual spaces

$$\langle i, k | j, k' \rangle = \delta_{ij} \delta_{kk'}, \quad (A6a)$$

$$\sum_{i=1}^N |i, k\rangle \langle i, k| = \hat{I}. \quad (A6b)$$

Using these relations, a straightforward calculation leads to the expression

$$\begin{aligned} \tilde{P}^{(k)}(s) &= \sum_{i=1}^N |i, k\rangle (s - \lambda_i^{(k)})^{-1} \\ &\quad \times \langle i, k | P^{(k)}(0) \rangle. \end{aligned} \quad (A7)$$

The inverse Laplace transform is easily performed and we obtain

$$\begin{aligned} P^{(k)}(t) &= \sum_{i=1}^N |i, k\rangle \langle i, k | P^{(k)}(0) \rangle \\ &\quad \times \exp(\lambda_i^{(k)} t). \end{aligned} \quad (A8)$$

Now let us call  $u_{nM+k, i}^{(k)}$  the  $n$ th component of the column vector  $|i, k\rangle$ . We easily obtain the expression for any  $P_{nM+k}(t)$  element of the distribution  $P(t)$

$$\begin{aligned} P_{nM+k}(t) &= \sum_i u_{nM+k, i}^{(k)} \langle i, k | P^{(k)}(0) \rangle \\ &\quad \times \exp(\lambda_i^{(k)} t). \end{aligned} \quad (A9)$$

We emphasize the practical interests of this method: The calculation is performed for any initial condition without the necessity of re-diagonalizing the matrices. The relative accuracy of the computed values is only dependent on the accuracy of the diagonalizations of the  $\hat{A}^{(k)}$  matrices. This remains constant whatever the given time  $t$  is when the distributions  $P_{nM+k}^{(k)}(t)$  are computed. In our case the relative accuracy of the diagonalizations is at least  $10^{-10}$ .

The method described above and the results obtained apply to any system of linear differential equations with time-independent coefficients. For the particular Eqs. (5)–(7) we consider in this paper, two features have some consequences on the long-time solution of Eqs. (5)–(7). First, there is a limit solution  $P^{(k)}(\infty)$  when  $t$  goes to infinity. This implies that in each matrix  $\hat{A}^{(k)}$  an eigenvalue, say  $\lambda_1^{(k)}$ , is zero and all others are negative. The limit solution is given by solving the system of algebraic equations

$$\hat{A}^k P^{(k)}(\infty) = 0 \quad (\text{A10})$$

or by the relation, from Eq. (A8),

$$P^{(k)}(\infty) = |1, k\rangle \langle 1, k | P^{(k)}(0) \rangle . \quad (\text{A11})$$

Also summing Eqs. (5)–(7) over  $n$  leads to the relation

$$\frac{d}{dt} \left[ \sum_n P_{nM+k}(t) \right] = 0 . \quad (\text{A12})$$

On each  $k$  block of probabilities the sum of probabilities is a constant. Since there are physical probabilities of counting  $n$  photons, we have the quantically induced relations

$$0 \leq \sum_{n=0} P_{nM+k}(t) \leq 1, \quad k=0, \dots, M-1 \quad (\text{A13a})$$

$$\sum_{n,k} P_{nM+k}(t) \equiv 1 . \quad (\text{A13b})$$

Using relations (A11)–(A13), we have

$$v_{1,j}^{(k)} = \frac{1}{N}, \quad l=1, N \quad (\text{A14})$$

when the  $v$  are the elements of the bra vector  $\langle i, k | :$

$$P^{(k)}(\infty) = \frac{1}{N} |i, k\rangle \sum_n P_{nM+k}(0) . \quad (\text{A15})$$

Equation (A15) has important physical implications in the case of the multiphoton amplifiers. We see that the long-time distribution is dependent on the eigenvectors of the matrix and on the sum of probabilities in each block  $k$  at initial time. So in the one-photon amplifier, as  $\sum_n P_n \equiv 1$ , the long-time distribution is only dependent on the eigenvectors i.e., the physical characteristics of the amplifier. On the contrary, in the multiphoton amplifiers the long-time solution is also dependent on the distribution of the photon probabilities in every block at time  $t=0$ . This can have important consequences when the input mean number of photons is very small. In particular, the amplifier noise, i.e., the field obtained by amplifying zero photon input, has nonzero probabilities in the sole  $k=0$  block.

## APPENDIX B: METHOD FOR COMPUTATION OF THE TIME INTENSITY CORRELATION FUNCTION

The second-order time correlation function can be written

$$\langle n_2, t + \tau; n_1, t \rangle = \sum_{n_1, n_2} n_2 P(n_2, t + \tau; n_1, t) n_1 P(n_1, t; \{P(0)\}, 0) , \quad (\text{B1})$$

where

$$P(n_2, t'; \{P(t)\}, t) \quad (\text{B2})$$

represents the conditional probability for counting  $n_2$  photons at time  $t' > t$  when the probability detection for the photons at time  $t (< t')$  is defined by the set of variables  $\{P(t)\}$ . In the expression (B1), in  $P(n_2, t + \tau; n_1, t)$ ,  $n_1$  means for a probability distribution

$$P_n(t) = \delta_{n, n_1} . \quad (\text{B3})$$

Using the fact that the Liouvillian equation is a first-order linear differential equation and referring to the quantities defined in Appendix A, expression (B1) can be rewritten

$$\langle n_2, t + \tau; n_1, t \rangle = \sum_{k=1}^N \sum_{i,j} \Theta_{ij}^k e^{\lambda_i^k \tau} e^{\lambda_j^k t} , \quad (\text{B4})$$

where

$$\Theta_{ij}^k = \sum_{n_1, n_2} (n_1 N + k)(n_2 N + k) u_{n_2, k, i}^{(k)} u_{n_1, k, j}^{(k)} V_{i, n_1, k}^{(k)} \langle j, k | P_0 \rangle . \quad (\text{B5})$$

We can easily write the normalized correlation function

$$\bar{g}_2(t, t + \tau) = \frac{\langle n_1, t + \tau; n_2, t \rangle - n_0 \langle n, t + \tau, \{P(0)\} \rangle}{\langle n_1, t; n_2, t \rangle - n_0 \langle n, t + \tau, \{P_0\} \rangle} , \quad (\text{B6})$$

TABLE I. Comparison of the computed  $t_{M\text{num}}$  time at which  $k_{n,M}(t)$  is maximum with the approximated values  $t_{M\text{cal}}$  obtained from Appendix C for  $M=1, 2, 3$ , and 4.

$M$	$\chi$	$k_n$	$\langle n \rangle_{\text{num}}$	$\langle n \rangle_{\text{cal}}$	$t_{M\text{num}}$	$t_{M\text{cal}}$
1	$7.1 \times 10^{-3}$	2	4.30	3.50	2.5	2.06
2	$5 \times 10^{-4}$	3	3.30	3.30	0.12	0.12
3	$2.9 \times 10^{-5}$	4	2.43	2.66	0.0075	0.025
4	$1.85 \times 10^{-6}$	5	2.05	2.56	0.0006	0.003

where  $\langle n, t + \tau, \{P_0\} \rangle$  is the mean number of photons at time  $t + \tau$ , given the initial conditions  $\{P_0\}$  and can be explicitly computed using formula (A8).

For a sufficiently long time  $t$ , formula (B4) can be simplified, using the fact that only the terms where  $\lambda_j^k = 0$  give a contribution. Assuming that in each block of the Liouvillian matrix this particular eigenvalue is indexed by 0, formula (B4) can be rewritten

$$\langle n_2, t + \tau; n_1, t \rangle = \sum_{k=1}^N \sum_1 \theta_{i0}^k e^{\lambda_i^k \tau}. \quad (\text{B7})$$

Except for very small values of  $\tau$  the evolution of the correlation function with  $\tau$  is governed by the smaller nonzero eigenvalue of each block  $k$ . So if  $N=1$ , the correlation time is directly given by

$$\tau_c = 1/\lambda_1^1, \quad (\text{B8})$$

where  $\lambda_1^1$  is the smallest of the nonzero eigenvalues, whereas if  $N > 1$  the correlation time cannot be simply defined and has no simple physical meaning.

#### APPENDIX C: APPROXIMATION METHOD TO CALCULATE THE TIME AT WHICH THE SECOND CUMULANT IS MAXIMUM

From Eq. (5) it is easy to calculate the derivative of the first moment given by

$$\langle n \rangle = \sum_n n P_M(n, t). \quad (\text{C1})$$

We find for  $\chi_M = 0$  and  $\beta_M = \beta$ ,

$$\frac{d\langle n \rangle}{M dt} = \left\langle \prod_{l=1}^M (n+l) \right\rangle - \beta \left\langle \prod_{l=0}^{M-1} (n-l) \right\rangle, \quad (\text{C2})$$

whose solution is simple for  $M=1$  but difficult to derive for  $M \geq 2$ .

For  $\chi_M \neq 0$  but small enough so that  $\chi_M n \ll 1$ , it can be shown

$$\frac{d\langle n \rangle}{M dt} \simeq \Delta \langle n^M \rangle - \chi_M \langle n^{2M} \rangle \quad (\text{C3})$$

with

$$\Delta = 1 - \beta.$$

Also by definition

$$\langle n^2 \rangle = k_{n,M} \langle n \rangle^2 + \langle n \rangle \simeq k_{n,M} \langle n \rangle^2. \quad (\text{C4})$$

On the other hand, we have noted that the maximum of  $k_{n,M}$  occurs for

$$\frac{dk_{n,M}}{dt} = 0$$

and

$$\frac{d^2\langle n \rangle}{dt^2} = 0$$

simultaneously. Making a new derivative of (C3) and taking into account (C4) we find

$$\langle n \rangle^M = \frac{\Delta}{2\chi_M k_{n,M}^M}. \quad (\text{C5})$$

Now we use our numerical results to verify the assumptions leading to (C5) by noting that  $k_{n,M} \sim M + 1$ . These values give reasonably good estimates of  $\langle n \rangle$  as we can see in Table I. However, calculation of  $t_M$  from the linear approximation of  $\langle n \rangle$  is clearly poor for  $M > 2$ .

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