PHOTON STATISTICS IN HARMONIC GENERATION PROCESSES WITH A WEAK INPUT CHAOTIC FIELD

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The time evolution of the normally ordered photon number variance of the fundamental mode in m-th harmonic generation processes in the case of a weak input chaotic field is considered. It is shown that the fundamental mode may reveal sub-Poissonian photon statistics, however, in the course of second-harmonic generation solely.

I. Introduction

Harmonics generation processes with an initially coherent fundamental mode yield fields exhibiting nonclassical effects of sub-Poissonian photon number statistics and squeezing [1–18]. As two mode phenomena they offer yet the possibility to study the intermodal correlations (for a review see [8]) and the validity of the Cauchy-Schwarz inequality [12].

To our best knowledge, Simaan and Loudon [19] have first shown explicitly for two-photon absorption that an initially chaotic field goes into a sub-Poissonian field. In turn, Trung and Schütte [20] and Peřinova and Peřina [21] have obtained for sum-frequency generation with chaotic input beams an enhancement of anticorrelation between the different field modes in comparison with the coherent case. It has been attributed to opposite fluctuations in the field modes [8].

Analytical solutions for the nonclassical effects mentioned above in generation processes have been found using the short-time approximation [1,2,4–18]. It is always interesting to present even an approximate analytical solution that is valid for all times. It is our aim to give an analytical approach to the problem of the photon number statistics of an initially weak chaotic fundamental mode in the course of m-th harmonic generation. Only for small photon numbers there exists a potential possibility of transformation of a chaotic fundamental mode into a sub-Poissonian one. The solution obtained will permit us to show explicitly that this transformation is, in fact, possible for second- (but not third- and higher-) harmonic generation.

II. Short-time solution for the fundamental mode in higher-harmonic generation

The rotating wave approximation Hamiltonian for the process of m-th harmonic generation reads

$$
\begin{aligned}
H &= H_{\text{free}} + H_{\text{int}}, \\
H_{\text{free}} &= \hbar\omega a^\dagger a + m\hbar\omega b^\dagger b, \\
H_{\text{int}} &= \hbar g_m \left(a^m b^\dagger + a^{\dagger m} b\right) .
\end{aligned} \tag{1}
$$

$a^\dagger(a)$ and $b^\dagger(b)$ are the photon creation (annihilation) operators for the fundamental and m-th harmonic modes, respectively. ω denotes the frequency of the fundamental field mode while g_m is the mode coupling. Owing to nonlinear coupling the operators a and b are functions of time not only by way of oscillating factors coming from the free evolution. We factor out the free evolution from the operators writing

$$
\begin{aligned}
a(t) &= a_s(t)\exp(-\mathrm{i}\omega t), \\
b(t) &= b_s(t)\exp(-\mathrm{i}m\omega t),
\end{aligned} \tag{2}
$$

where $a_s(t)$ and $b_s(t)$ are slowly varying parts of the operators and satisfy the same commutation rules as the operators a and b:

$$
[a, a^\dagger] = [b, b^\dagger] = 1 . \tag{3}
$$

The Hamiltonian (1) leads to the following coupled equations of motion:

$$
\begin{aligned}
\dot{a}_s &= -\mathrm{i}mg_m a_s^{\dagger\, m-1} b_s, \\
\dot{b}_s &= -\mathrm{i}g_m a_s^m .
\end{aligned} \tag{4}
$$

The long-time evolution of the process will be found by performing numerical calculations with the method of diagonalization of the interaction Hamiltonian (1), the details of which are described elsewhere [22]. To start with, however, let us briefly recall the short-time solution for the fundamental mode. This solution is related with the expansion of the operators in a Taylor series around $t = 0$, where t is the time which it takes for the beams to traverse the medium. Here, we are interested in the time evolution of the fundamental beam. The solution for the operator a_s within an accuracy of t^2 is [11]

$$
\begin{aligned}
a_s(t) &= a_0 - \mathrm{i}mg_m t a_0^{\dagger\, m-1} b_0 + \frac{m}{2} g_m^2 t^2 \left[\sum_{r=1}^{m-1} r! \binom{m-1}{r} \binom{m}{r} \right. \\
&\quad \left. \times\, [: n_{f0}^{m-1-r} :] a_0 n_{m0} - [: n_{f0}^{m-1} :] a_0 \right] + \ldots ,
\end{aligned} \tag{5}
$$

where the symbol : : stands for normal ordering of the photon creation and annihilation operators and the subscript 0 denotes that the operators (in what follows, other quantities as well) are taken at $t = 0$. n_{f0} and n_{m0} are the photon-number operators for the fundamental and harmonic mode, respectively.

The one-time first- and second-order field correlation functions are defined by

$$\begin{aligned} G^{(1)}(t) &= \langle n(t) \rangle , \\ G^{(2)}(t) &= \langle n(t)^2 \rangle - G^{(1)}(t) . \end{aligned} \tag{6}$$

The symbol $\langle \quad \rangle$ denotes quantum-mechanical averaging over the states of the field. The harmonic field is in a vacuum state at $t = 0$, so that the following initial conditions are satisfied $\langle 0|b_0|0\rangle = \langle 0|n_{m0}|0\rangle = 0$. In order to obtain the field correlation function for the fundamental mode being initially in a coherent or chaotic state one has to calculate the quantum-mechanical expectation values in Eqs. (6) by summing up the expressions for $n(t) = a_s^\dagger(t)a_s(t)$ and $n^2(t) = (a_s^\dagger(t)a_s(t))^2$ over the Poissonian or geometrical photon number distributions P_n, respectively:

$$P_n = \exp(-\bar{n}_{f0})\frac{\bar{n}_{f0}^n}{n!} , \tag{7}$$

$$P_n = \frac{\bar{n}_{f0}^n}{(\bar{n}_{f0}+1)^{n+1}} , \tag{8}$$

where $\bar{n}_{f0} = \langle a_0^\dagger a_0 \rangle$ is the initial mean photon number.

On insertion of Eq. (5) into the definitions (6), after some algebra one finds

$$\begin{aligned} G_f^{(1)}(t) &= \bar{n}_{f0} - m g_m^2 t^2 G_{f0}^{(m)} + \ldots , \\ G_f^{(2)}(t) &= G_{f0}^{(2)} - m g_m^2 t^2 \left[2G_{f0}^{(m+1)} + (m-1)G_{f0}^{(m)} \right] + \ldots , \end{aligned} \tag{9}$$

where

$$G_{f0}^{(m)} = \sum_{n=m} P_n \frac{n!}{(n-m)!} . \tag{10}$$

Beginning the summation from $n = m$ reflects the fact that at least m photons are needed to have a nonzero m-th order factorial moment.

By definition, the normally ordered photon number variance is

$$V(t) = G^{(2)}(t) - \left[G^{(1)}(t) \right]^2 . \tag{11}$$

Negative values of the normally ordered variance ($V < 0$) indicate sub-Poissonian photon number statistics.

For the fundamental mode Eq. (11) takes the form

$$V_f(t) = V_{f0} - m g_m^2 t^2 \left\{ 2\left[G_{f0}^{(m+1)} - G_{f0}^{(m)} G_{f0}^{(1)} \right] + (m-1)G_{f0}^{(m)} \right\} , \tag{12}$$

and if the field is initially coherent, the above expression transforms to [8]

$$V_f(t) = -m(m-1) g_m^2 t^2 \bar{n}_{f0}^m , \tag{13}$$

pointing directly to sub-Poissonian photon statistics at the onset of the interaction. The variance becomes negative more steeply as m and $\bar{n}$ increase. For an initially chaotic fundamental mode from Eq. (12), we get

$$V_f(t) = \bar{n}_{f0}^2 - mm!g_m^2 t^2 \bar{n}_{f0}^m (2m\bar{n}_{f0} + m - 1) . \tag{14}$$

The variance, as expected, starts to decrease immediately after switching on the interaction and this tendency is more rapid as m and $\bar{n}$ grow. However, the conclusion that for a given, sufficiently large m and $\bar{n}$, the variance could become negative at an appropriate time t would be too far reaching. Equation (14) describes the very early stage of the process and, in fact, the transformation into a sub-Poissonian field occurs for small $\bar{n}_{f0}$ and, as we will see further on, for $m = 2$ only), and it corresponds to the general feature of this quantum effect that it is meaningful for not very large photon numbers.

III. Analytical solution

We intend to find an approximate analytical solution to the problem discussed, permitting us to show explicitly that only in the course of second-harmonic generation the fundamental mode may exhibit sub-Poissonian photon number statistics. To obtain this solution, we consider the multiphoton Jaynes-Cummings model (JCM) with an initially unexcited two-level atom, described at exact m-photon resonance and in rotating wave approximation by the following effective Hamiltonian:

$$\begin{aligned} H &= H_{\text{free}} + H_{\text{int}} , \\ H_{\text{free}} &= m\hbar\omega S^3 + \hbar\omega a^\dagger a , \\ H_{\text{int}} &= \hbar g_m [a^{\dagger m} S^- + a^m S^+] , \end{aligned} \tag{15}$$

where ω denotes the frequency of the field mode while g_m is now the multiphoton atom-field coupling. S^-, S^+ and S^3 are the pseudospin lowering, raising and inversion operators of the atom, respectively, and

$$[S^-, S^+] = -2S^3 . \tag{16}$$

For the multiphoton JCM's ($m \geq 2$) the basic effective Hamiltonian (15) omits the dynamic Stark shifts of the upper and lower atomic levels due to transitions to intermediate states and its validity has been questioned (e.g. [23–30]). We would like to stress that we are not interested here in the time behaviour of the multiphoton JCM but only in the application of the above Hamiltonian to the description of the time evolution of the fundamental mode in m-th harmonic generation.

From the second commutation relation (3) and the relation (16) it arises that the interaction Hamiltonians (1) and (15) lead to almost the same results if the average value of the atomic inversion $\langle S^3(t) \rangle$ remains close to $-1/2$. Obviously, it takes place at the beginning of the interaction independently of the initial mean photon number. On the other hand, the same situation occurs for an arbitrary time at sufficiently weak initially coherent or chaotic fields $\bar{n} < 1$.

Let the field be initially in a photon number state $|n\rangle$. The atom starts in its lower state $|-\rangle$. Then, the interaction-picture state of the system reads

$$|\Psi(t)\rangle = |-, n\rangle C^{(n)}_{-m}(t) + |+, n-m\rangle C^{(n)}_{+m}(t)\,, \tag{17}$$

where $|+\rangle$ denotes the upper state of the atom and the probability amplitudes $C^{(n)}_{-m}(t)$ and $C^{(n)}_{+m}(t)$ are

$$\begin{aligned} C^{(n)}_{+m}(t) &= -\mathrm{i}\sin[\Omega_m(n)t], \\ C^{n}_{-m}(t) &= \cos[\Omega_m(n)t]\,. \end{aligned} \tag{18}$$

$\Omega_m(n)$ is the quantum Rabi frequency describing oscillations of the atom-field system

$$\Omega_m(n) = g_m\sqrt{\frac{n!}{(n-m)!}}\,. \tag{19}$$

To have this frequency nonzero n should be $\geq m$.

From the definitions (6), we have

$$\begin{aligned} {}^{n}G^{(1)}(t) &= \langle\Psi(t)|a^\dagger a|\Psi(t)\rangle = n - m|C^{(n)}_{+m}(t)|^2\,, \\ {}^{n}G^{(2)}(t) &= \langle\Psi(t)|a^\dagger a^\dagger aa|\Psi(t)\rangle = n^2 - n + m(m+1-2n)|C^{(n)}_{+m}(t)|^2\,. \end{aligned} \tag{20}$$

The superscript n preceding the correlation functions will denote that the field is initially in a Fock state.

In turn, the atomic inversion reads

$${}^{n}\langle S^3(t)\rangle = -\frac{1}{2} + |C^{(n)}_{+m}(t)|^2\,. \tag{21}$$

Obviously, for a Fock field, the atomic inversion periodically reaches its maxima (1/2) and minima (−1/2) independently of n (certainly, n should be $\geq m$); its quasi stationary value is equal to zero.

For an initially coherent or chaotic field from (20) we simply get

$$\begin{aligned} G^{(1)}(t) &= \sum_{n=0}^{\infty} P_n {}^{n}G^{(1)}(t) = \bar{n} - m\sum_{n=m}^{\infty} P_n|C^{(n)}_{+m}(t)|^2\,, \\ G^{(2)}(t) &= \sum_{n=0}^{\infty} P_n {}^{n}G^{(2)}(t) = G^{(2)}_0 + m(m+1)\sum_{n=m}^{\infty} P_n|C^{(n)}_{+m}(t)|^2 \\ &\quad - 2m\sum_{n=m}^{\infty} P_n n|C^{(n)}_{+m}(t)|^2\,, \end{aligned} \tag{22}$$

while the atomic inversion then evolves according to

$$\langle S^3(t)\rangle = -\frac{1}{2} + \sum_{n=m}^{\infty} P_n|C^{(n)}_{+m}(t)|^2\,. \tag{23}$$

Owing to the form of the Rabi frequency (19) the summation over n in the above equations starts from $n = m$ (in the terms containing the probability amplitude $C_{+m}^{(n)}(t)$).

Let us expand the amplitude $C_{+m}^{(n)}(t)$ in a power series in t. Within an accuracy of t^2 we arrive at

$$\begin{aligned} \sum_{n=m}^{\infty} P_n |C_{+m}^{(n)}(t)|^2 &= g_m^2 t^2 \sum_{n=m}^{\infty} P_n \frac{n!}{(n-m)!} = g_m^2 t^2 G_0^{(m)} , \\ \sum_{n=m}^{\infty} P_n n |C_{+m}^{(n)}(t)|^2 &= g_m^2 t^2 \sum_{n=m}^{\infty} P_n n \frac{n!}{(n-m)!} = g_m^2 t^2 [G_0^{(m+1)} + m G_0^{(m)}] . \end{aligned} \quad (24)$$

Hence, for the normally ordered photon number variance (11) within an accuracy of t^2, we arrive as expected, at the solution identical with (14) for the fundamental mode in the course of m-th harmonic generation.

The quasi stationary value of the atomic inversion (23) is equal to

$$\langle S^3 \rangle_{\mathrm{qs}} = -\frac{1}{2} \sum_{n=0}^{m-1} P_n . \quad (25)$$

Obviously, for strong coherent and chaotic fields it is approximately equal to zero. This case is beyond the scope of our considerations.

In turn, for a chaotic field this inversion after simple summation takes the form

$$\langle S^3 \rangle_{\mathrm{qs}} = -\frac{1}{2}(1 - q^m) , \quad (26)$$

where

$$q = \frac{\bar{n}}{\bar{n} + 1} . \quad (27)$$

If the chaotic field is weak ($q^m \ll 1$), the system oscillates with small amplitudes around the value (26) and the temporary values of $\langle S^3(t) \rangle$ then remain close to $-1/2$. Hence, it is reasonable to describe the time evolution of an initially weak chaotic fundamental mode (coherent as well) in higher-harmonic generation with the help of the multiphoton JCM. Obviously, the case of weak input fields is less interesting from the experimental point of view. However, it is desired for the completeness of our knowledge about quantum features of harmonic generation processes caused, in particular, by chaotic fields.

The first-order correlation function (20) may be presented as follows:

$$G^{(1)}(t) = \bar{n} - \frac{m}{2} [C - W_0(t)] , \quad (28)$$

where with respect to Eqs. (25) and (26) the constant term C reads

$$C = \sum_{n=m}^{\infty} P_n = 1 + 2\langle S^3 \rangle_{\mathrm{qs}} = q^m , \quad (29)$$

while the function $W_0(t)$ has the form

$$W_0(t) = (1-q)\sum_{n=m}^{\infty} q^n \cos\left[2\Omega_m(n)t\right] . \tag{30}$$

To calculate the function $W_0(t)$ we use a linear approximation in n for the Rabi frequency in the range $n \geq m$ as it was done for the standard JCM ($m=1$) [31][1]) with an initially unexcited atom or for a multiphoton micromaser case [32]. Quite generally one can write

$$2\Omega_m(n) = A_{0m} + (n-m)A_{1m} . \tag{31}$$

We assume that the above equation is exact in the first two points $n=m$ and $n=m+1$:

$$2\Omega_m(m) = A_{0m}, \qquad 2\Omega_m(m+1) = A_{0m} + A_{1m} . \tag{32}$$

From the definition (19) we find that

$$A_{0m} = 2g_m\sqrt{m!}, \qquad A_{1m} = A_{0m}(\sqrt{m+1}-1) . \tag{33}$$

Equation (30) then contains an easily performed summation of the geometrical progression

$$W_0(t) = \frac{q^m}{2}(1-q)\exp(\mathrm{i}A_{0m}t)\sum_{k=0}^{\infty} q^k \exp(\mathrm{i}kA_{1m}t) + \mathrm{c.c.}, \tag{34}$$

and finally one arrives at

$$G^{(1)}(t) = \bar{n} - \frac{m}{2}q^m\left[1-(1-q)\frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}}\right], \tag{35}$$

where

$$\begin{gathered} D(\tau) = 1+q^2-2q\cos\tau, \\ \cos\phi(\tau) = \frac{1-q\cos\tau}{\sqrt{D(\tau)}}, \qquad \sin\phi(\tau) = \frac{q\sin\tau}{\sqrt{D(\tau)}}, \end{gathered} \tag{36}$$

and the two time scales T and τ read

$$T = A_{0m}t, \qquad \tau = A_{1m}t \tag{37}$$

and they determine "fast oscillations" of the type $\cos T$ and a "slow envelope" dependent on τ, respectively.

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[1]) The original version of this paper contained the results for the multiphoton JCM as well.

In turn, the second-order field correlation function may be written in the form

$$G^{(2)}(t) = 2\bar{n}^2 + \frac{m(m+1)}{2}\left[C - W_0(t)\right] - m\left[\sum_{n=m}^{\infty} nP_n - W_1(t)\right]. \tag{38}$$

The function $W_1(t)$ reads

$$\begin{aligned} W_1(t) &= q(1-q)\frac{\partial}{\partial q}\left(q^m \frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}}\right) = \\ &= (m-1)W_0(t) + q^m(1-q)\frac{\cos[T+2\phi(\tau)]}{\sqrt{D(\tau)}}. \end{aligned} \tag{39}$$

Hence, the approximate second-order correlation function is found to be

$$\begin{aligned} G^{(2)}(t) &= 2\bar{n}^2 - mq^m\left\{\bar{n} + \frac{m-1}{2} - (1-q)\left[\frac{m-3}{2}\frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}}\right.\right. \\ &\quad \left.\left. + \frac{\cos[T+2\phi(\tau)]}{D(\tau)}\right]\right\}. \end{aligned} \tag{40}$$

For the normally ordered variance, from Eqs. (35) and (40), one gets

$$\begin{aligned} V(t) &= \bar{n}^2 - mq^m\left\{\frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}} - (1-q)\frac{\cos[T+2\phi(\tau)]}{D(\tau)} + \left[1-(1-q)\right.\right. \\ &\quad \left.\left. \times\frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}}\right]\left[\frac{m-1}{2} + \frac{m}{4}q^m\left(1-(1-q)\frac{\cos[T+\phi(\tau)]}{\sqrt{D(\tau)}}\right)\right]\right\}. \end{aligned} \tag{41}$$

We postulate that the results (35), (40) and (41) describe the time behaviour of the fundamental mode in m-th harmonic generation when at $t = 0$ the fundamental mode is chaotic and weak ($\bar{n}_{f0} < 1$).

IV. Discussion

Figure 1 shows the time evolution of the exact, numerically computed, mean photon number of the initially chaotic fundamental mode and of the approximate photon number (35) in third-harmonic generation. Agreement between these two curves is really satisfactory, at least for the photon numbers assumed.

Let us estimate very roughly the signs at the minimal values of $V(t)$. We neglect in the square bracket (41) all the terms containing q and its higher powers. On this assumption we arrive at

$$V \simeq \bar{n}^2 - \frac{1}{2}m(m-1)q^m(1-\cos T). \tag{42}$$

Putting furthermore $\cos T = -1$ we minimize the value (42) of V. We then have

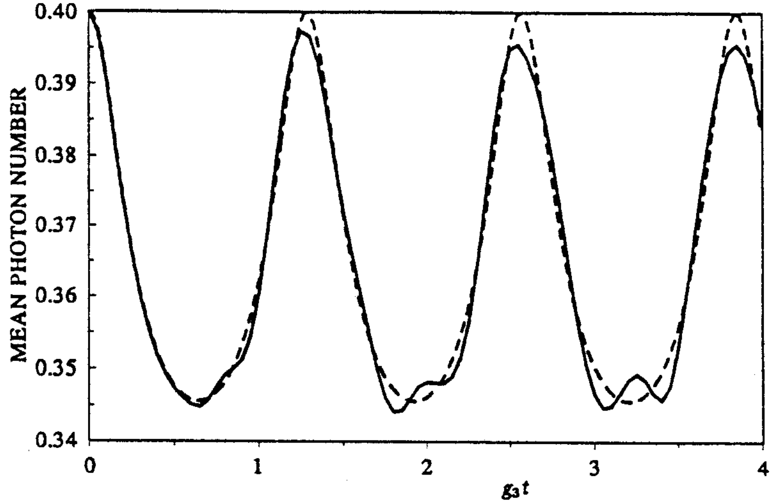


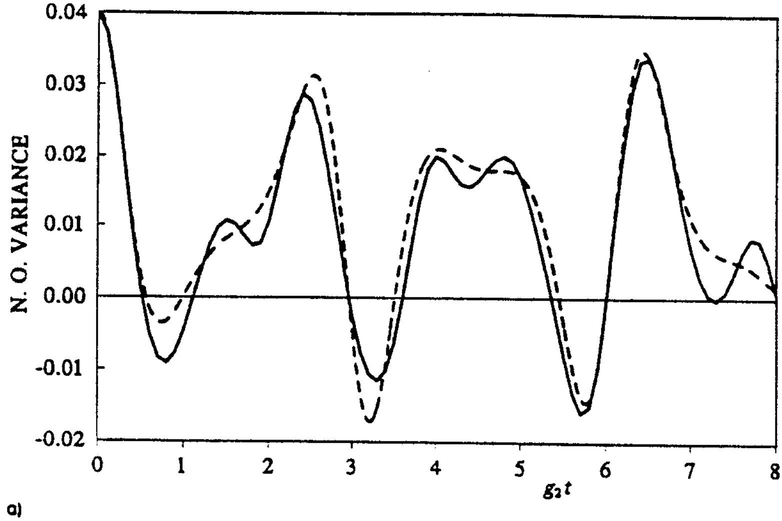
Fig. 1. Time evolution of the mean photon number of the initially chaotic fundamental in third-harmonic generation for $\bar{n}_{f0} = 0.4$. Solid line corresponds to the exact computer solution while dashed line corresponds to the approximate result (35).

$$V \simeq \frac{\bar{n}^2}{(\bar{n}+1)^m}\left[(\bar{n}+1)^m - m(m-1)\bar{n}^{m-2}\right] . \tag{43}$$

For second-harmonic generation this quantity may take negative values for $\bar{n} < 0.41$. The bound on sub-Poissonian photon statistics estimated in such a rough manner, additionally from the approximated formula, does not remain in good agreement with the limit found numerically: $\bar{n} < 1.2$ (for times here assumed $gt \leq 8$). In turn, for $m \geq 3$ the term in the square bracket of Eq. (43) is always positive and sub-Poissonian photon number statistics cannot be observed in these processes in the fundamental beam when it is chaotic at the input to the medium. Second-harmonic generation differs from other harmonic generation processes in this respect. Figures 2a and b illustrate this situation.

Figure 3, representing the computer solution for the fundamental beam in the course of second-harmonic generation, shows how the widths of the photon number intervals in which sub-Poissonian photon statistics appears vary in time. For the clarity of the graph only the non-negative values of the quantity $(-V)$ are plotted. Therefore the positive values over the plane $(gt, \bar{n})$ immediately point to sub-Poissonian photon statistics. From this figure the exact, earlier mentioned, limit on sub-Poissonian photon statistics is evident. A rather interesting feature of the effect in question is seen from the first "hummock"; sub-Poissonian photon statistics starts to appear earlier as $\bar{n}$ grows.

Similarity of the description of the time evolution of the multiphoton JCM and harmonic generation processes turned our attention to the possibility of obtaining the sub-Poissonian fundamental mode in second-harmonic generation even in the



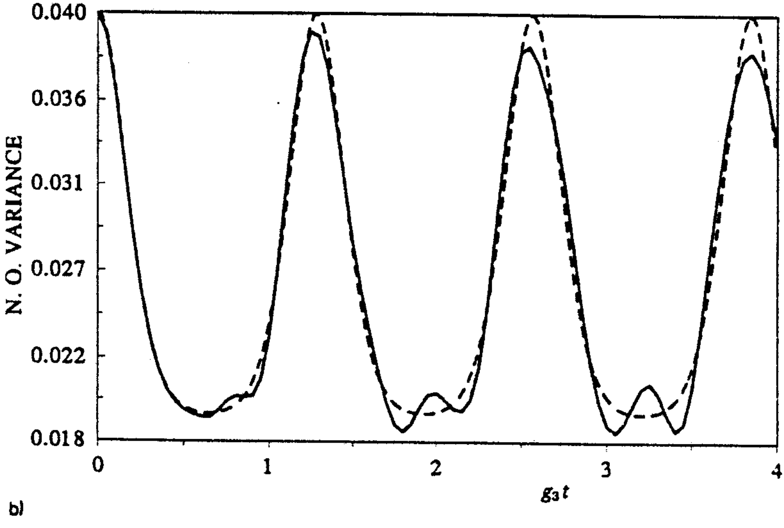


Fig. 2. Oscillations of the exact normally ordered photon number variance (solid line) and of the approximate one (41) (dashed line) for the initially chaotic fundamental in a) second-harmonic and b) third-harmonic generation; $\bar{n}_{f0} = 0.2$.

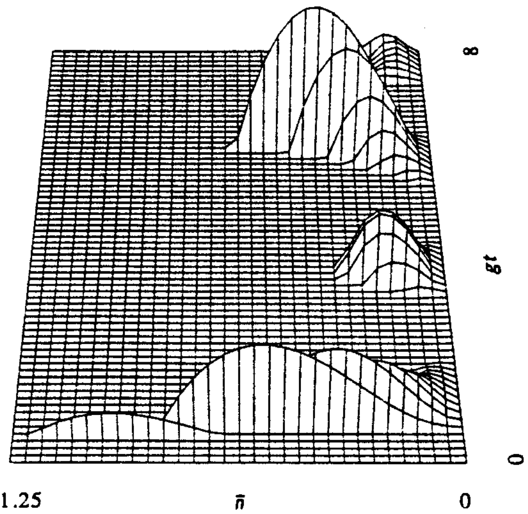


Fig. 3. Non-negative values of $(-V)$ versus dimensionless evolution time gt and $\bar{n}$ in second-harmonic generation (computer solution).

case when the input radiation is chaotic. On one hand, we are well aware that the bound on the number of the incident photons for the presence of this statistics creates a serious restriction as for the observation of second-harmonic generation. On the other hand, sub-Poissonian photon statistics is just meaningful for relatively small photon numbers causing a given nonlinear optical process: the deviation of the second-order coherence degree from unity is proportional to $1/\bar{n}$. In the case of coherent input the sub-Poissonian photon statistics is always present in generation processes, independently of $\bar{n}$. The only problem is then how to reconcile the efficiency of the process with the magnitude of sub-Poissonian photon statistics. To conclude, the result here obtained, describing the non-classical behaviour of the field statistics in second-harmonic generation with the chaotic input, although less useful from the experimental point of view, supplements our theoretical knowledge about quantum features of higher-harmonic generation processes.

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