SUB-POISSONIAN LIGHT IN THIRD-HARMONIC GENERATION: QUANTUM PREDICTIONS VIA CLASSICAL TRAJECTORIES

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Sub-Poissonian light in the third-harmonic generation process is studied numerically and analytically. Special regime exhibiting the time-stable maximum sub-Poissonian behaviour with the Fano factor $F \approx 0.81$ is found and analyzed. Theoretical prediction of the Fano factor and explanation of the extraordinary time stability of the sub-Poissonian behaviour are given using the semiclassical method of classical trajectories.

1 Introduction

The second- and higher-harmonic generation processes offer a very effective method for the production of strong coherent light with shorter wavelength (UV and X light) [1] and for the generation of light with more controlled level of noise (the squeezed, antibunched or sub-Poissonian light).

The light with photocount-noise level smaller than the shot-noise limit is called the sub-Poissonian light. Such optical field has obviously better signal-to-noise ratio than any classical light and can have many applications in, e.g., precise photometry, spectroscopy or optical communications. In experiments, the Fano factor $F = \mathrm{Var}(n) / \langle n \rangle$ is often used for a simple description of the photon-number statistics. Here, $\langle n \rangle$ represents the mean number of detected photons and $\mathrm{Var}(n)$ is its variance. Since the shot-noise limit is obtained by detection of coherent light (satisfying $\mathrm{Var}(n_{\mathrm{coh}}) = \langle n_{\mathrm{coh}} \rangle$), an optical field is sub-Poissonian if $F < F_{\mathrm{coh}} = 1$. Such kind of light has no classical analogue and can be properly described by quantum optics only. Since the classic experiment of Short and Mandel [2], sub-Poissonian light has been generated and observed in many laboratories [3].

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Recently, the third harmonic generation (THG) has also attracted a considerable interest as an effective material-diagnostic tool (see, e.g., [4]). THG, in comparison with second-harmonic generation, is generally a weak process but, being dipole allowed, occurs in all materials, including dielectric materials with inversion symmetry.

In the previous paper [5] we studied the second-harmonic generation process. We proposed the optimal method to generate light with the smallest photon-number noise and specified the conditions, under which the production of this noise-reduced light is the most stable in time. It was shown that the best results are obtained for the fundamental and second-harmonic coherent inputs in phase and with the amplitude ratio $2:1$. Under this initial condition, the second-harmonic output is sub-Poissonian with the Fano factor $F = 5/6 = 0.83$ (i.e., 0.79 dB below the shot-noise limit) for wide ranges of interaction lengths and input intensities. The above conclusion was proved numerically by solving the Schrödinger equation and confirmed analytically by applying the semiclassical method of classical trajectories.

In the present paper we will study the third harmonic (and subharmonic) generation process and its photon statistics. We shall see that this process is similar in many aspects to the second-harmonic generation and analogous methods can be used for solving it. We shall show, as our main result, that the lowest-value time-stable Fano factor of the third-harmonic output mode is $F = 13/16$ and the observed noise should be 0.90 dB below the shot-noise limit. This low-level noise is nearly independent of the interaction length and input intensities by assuming only that the relative amplitudes are in the optimal ratio 3:1 and the coherent input beams are in phase.

In quantum optics, the third-harmonic generation or three-photon down conversion (third-subharmonic generation) are described by the interaction Hamiltonian

$$H = \hbar g \left(a_1^3 a_3^\dagger + a_1^{\dagger 3} a_3 \right) , \tag{1}$$

where a_1 and a_3 denote annihilation operators of the fundamental and third-harmonic modes, respectively, and g is a nonlinear coupling parameter. Since no exact solution of quantum dynamics of the model (1) can be found, some appropriate analytical approximations or numerical methods have to be used for describing the conversion efficiency, quantum noise statistics or other characteristics of the process [6,7]. Here, we apply several methods including (i) short-time quantum expansions, (ii) numerical solution of the Schrödinger equation, (iii) strong-field classical solutions, and (iv) classical trajectory method.

2 Quantum analysis

The first predictions of sub-Poissonian photon-number statistics in second- or higher-harmonic generations were obtained under the short-time ($gt \ll 1$) approximation (see, e.g., [6,8]). For the coherent complex inputs $\alpha_1 = r_1 \exp(\mathrm{i}\phi_1)$ and $\alpha_3 = r_3 \exp(\mathrm{i}\phi_3)$, the short-time evolutions of the Fano factors are given by (see, e.g.,[10])

$$F_1 = 1 - 12gtr_1r_3\sin\theta + \cdots,$$
$$F_3 = 1 - 36\,(gt)^3\, r_1^3\,(r_1^2+2)\, r_3\sin\theta + \cdots. \tag{2}$$

The initial phase difference $\theta = 3\phi_1 - \phi_3$ determines (i) whether Eq. (2) describes the harmonic generation ($\omega+\omega+\omega \to 3\omega$) or the reversed process of subharmonic generation ($3\omega \to \omega+\omega+\omega$), and (ii) whether the sub-Poissonian or super-Poissonian light is generated in the first stage of the interaction. For example, in the case of $\sin\theta > 0$, the photon statistics of both generated outputs are sub-Poissonian, $F_{1,3} < 1$.

In the spontaneous short-time THG process ($\alpha_3 = 0$), the sub-Poissonian light is generated as described by the solutions [10]

$$F_1 = 1 - 6\,(gt)^2\, r_1^4 + \cdots,$$
$$F_3 = 1 - 9\,(gt)^4\, r_1^6\,(r_1^2+2) + \cdots. \tag{3}$$

On the contrary, the sub-Poissonian light is not observed ($F_{1,3} > 1$) in the reversed spontaneous process, i.e., for $\alpha_1 = 0$. Here, the subfrequency mode starts at $t = +0$ with super-Poissonian statistics ($F_1 = 3$) as being composed only from triplets of the subfrequency photons produced by the decay process of the pump photons, i.e., $3\omega \to \omega+\omega+\omega$.

For longer interaction times (when the short-time condition, $gt \ll 1$, is not fulfilled) no analytical predictions exist and numerical methods have to be applied. We use two numerical methods to study the long-time quantum dynamics: (i) the well-known method of diagonalization of Hamiltonian (1) originally used in Ref. 11 and (ii) the method of global characteristics based on the analysis of spectra of the density matrix [12]. We obtain the following results: for random choice of initial states α_1 and α_3, the evolution usually exhibits, after relaxation during initial short period of time (for $gt \gtrsim 1$), a strongly super-Poissonian behaviour ($F_{1,3} \gg 1$) in both modes. The only exception is a certain set of initial states concentrated along the line $\alpha_1 = 3r, \alpha_3 = r$, $r > 0$ (see Fig. 1) for the phase mismatch $\theta = 0$. It is worth noting that the zero mismatch is required in this case, contrary to the out-of-phase mismatch ($\theta = \pi/2$) needed to achieve the maximum sub-Poissonian behaviour in the short-time limit.

Instead of standard time evolution plots, the global Fano-factor characteristics F_{3G} [12] seem to be the best representation of the long-time sub-Poissonian behaviour of the third harmonic mode. In Fig. 1, we plot the global Fano factors in their dependence on the input coherent amplitudes of the fundamental (α_1) and third-harmonic (α_3) modes. By analyzing Fig. 2, we find that the global Fano factor F_{3G} of the harmonic mode remains constant at the value $F_3 = 0.81 < 1$ along the line $\alpha_1 = 3r$, $\alpha_3 = r$ for a wide range of amplitudes r but not close to the origin, i.e. $r > 1$.

For comparison, we study the long-time dynamics by applying the standard quantum method of diagonalization of Hamiltonian (1). Our quantum analysis leads

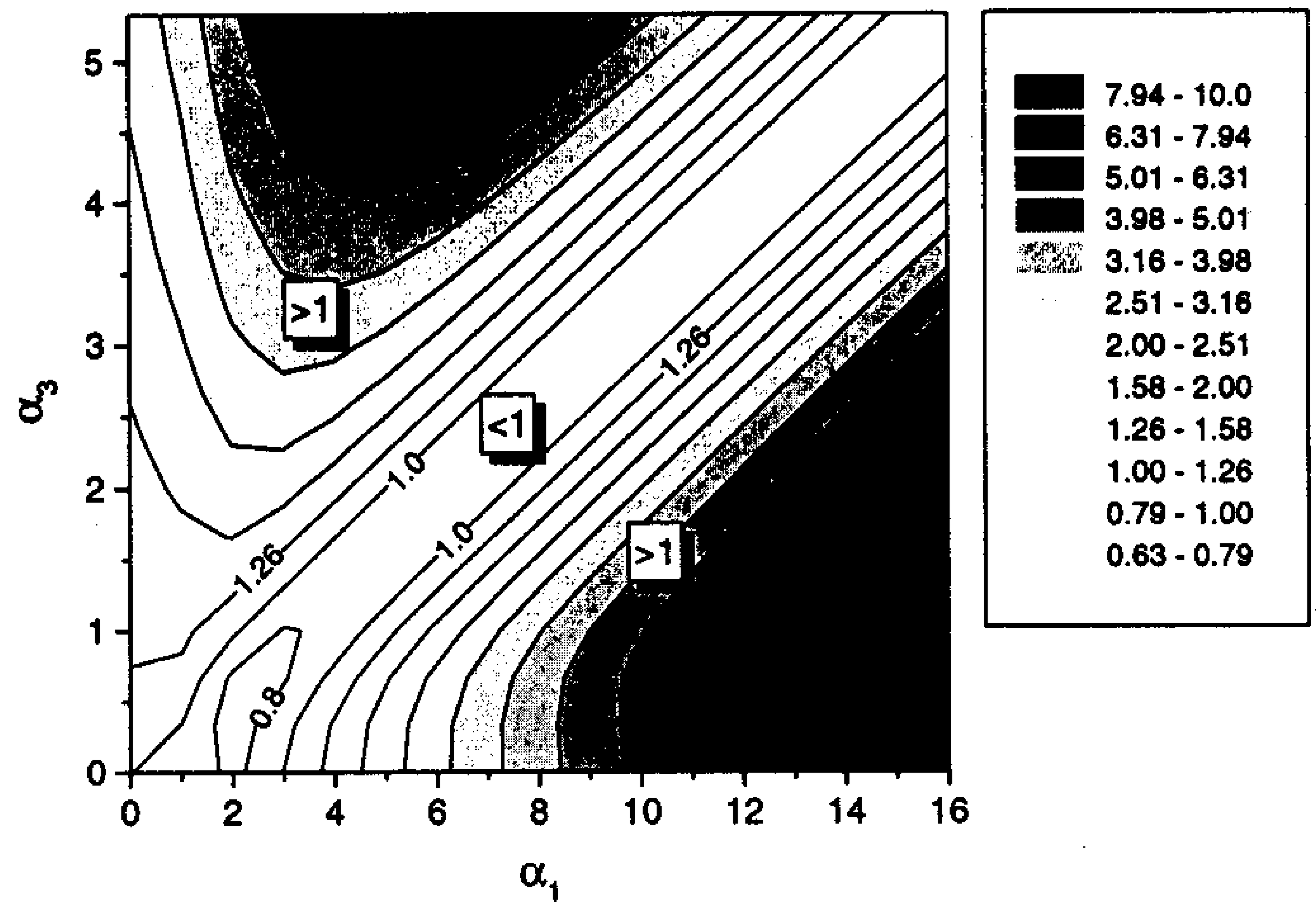

Fig. 1. Global Fano factor F_{3G} of the third-harmonic mode in the plane of initial coherent real ($\theta = 0$) amplitudes α_1 and α_3. Labels denote regions of the globally sub-Poissonian, Poissonian and super-Poissonian dynamics.

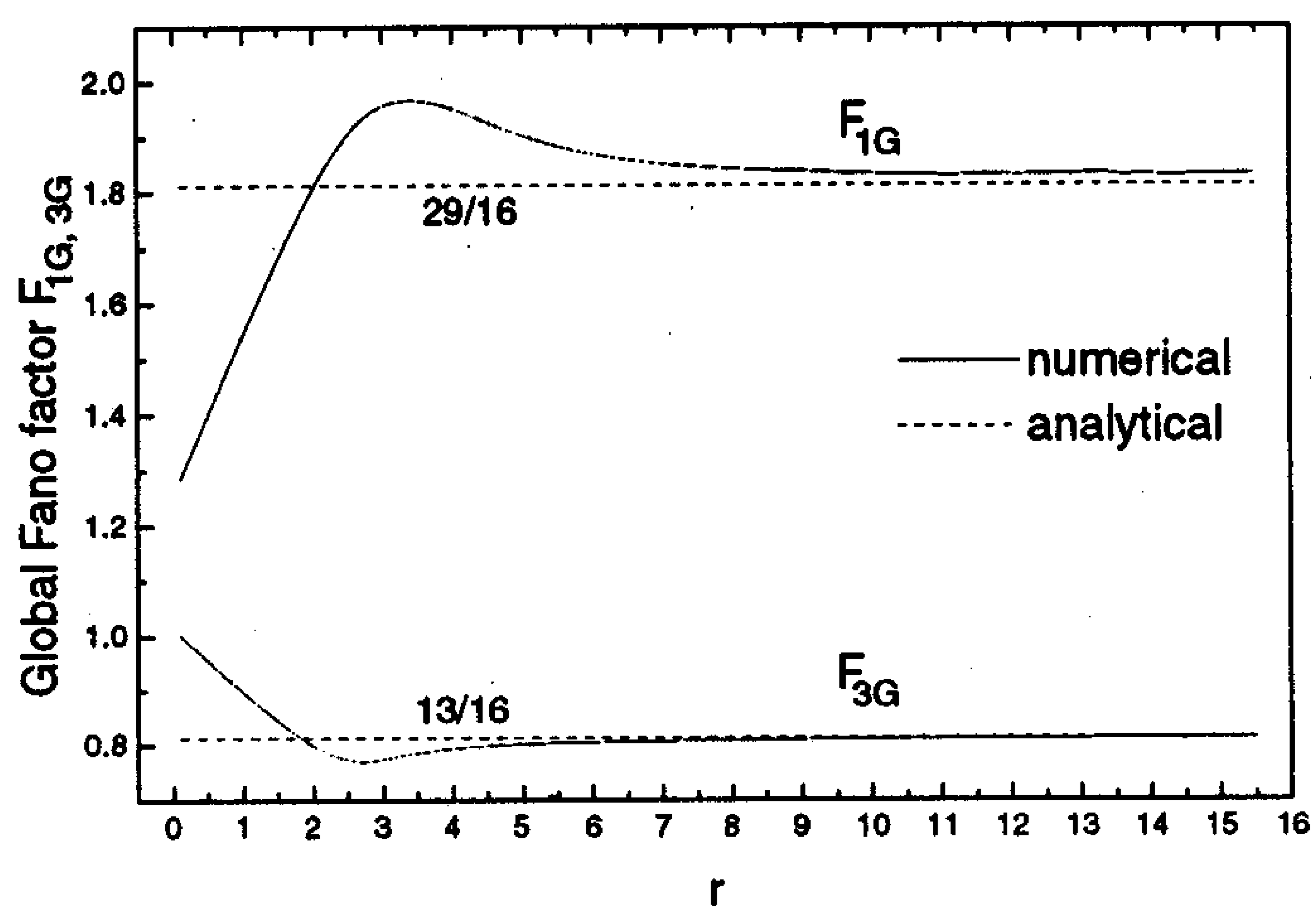

Fig. 2. Global Fano factors of the fundamental (F_{1G}) and third-harmonic (F_{3G}) modes for initial states $\alpha_1 = r$ and $\alpha_3 = r/3$, with $\theta = 0$. Solid curves represent the numerical results obtained by solving the Schrdinger equation, and dashed lines are the analytical results given by the method of classical trajectories.

to the conclusion that, under the above initial conditions, the time-stable sub-Poissonian behaviour is observed with the Fano factor F_3 approximately the same as the global Fano factor F_{3G}. Example of some typical time evolution is given in Fig. 3. One can see that the harmonic mode, after settling-down from its initial relaxation oscillations, remains sub-Poissonian independently of the interaction time. In the classical theory of THG this is called the *no energy transfer* [13] since in this case

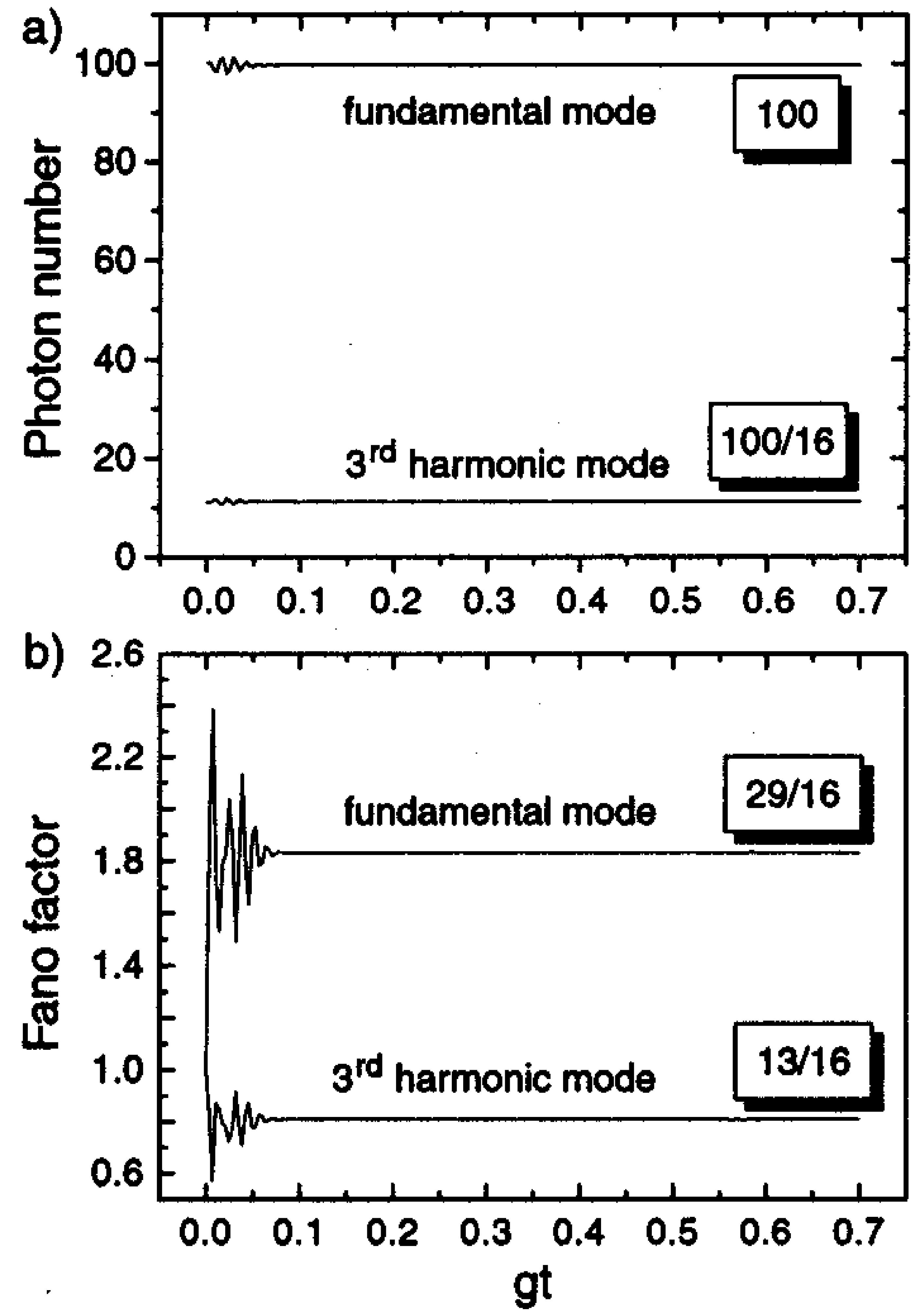

Fig. 3. Time evolution of a) mean photon numbers and b) Fano factors in the fundamental and third-harmonic modes for the no-energy-transfer case, i.e., for the initial conditions $\alpha_1 = 10$ and $\alpha_3 = 10/3$.

(and only here) the energy and photon numbers of both modes are conserved in time, and $n_1(t) = 9r^2 = \text{const}$ and $n_3(t) = r^2 = \text{const}$ holds. So, a question arises why in this case the output photon-number statistics remains sub-Poissonian in the harmonic mode and weakly super-Poissonian in the fundamental mode with nearly constant Fano factors $F_1 = 1.81$ and $F_3 = 0.81$? [1]) In what follows we will try to elucidate this behaviour.

First, we can calculate the Husimi Q-functions, as we did in our previous paper [5]. In the *no-energy-transfer* case, one could observe that the cross sections of both the Husimi Q-functions evolve from small circles centered at $\alpha_{1,3}$ to a rotated banana (or crescent) shapes and, finally, to ring shapes. The observed time stability of the photon-number statistics can be explained as a result of the stability of the Q-function cross-section rings. The evolution proceeds exactly in the same manner as in the case of the second-harmonic generation discussed in detail in Ref. 5. Thus, for brevity, we do not present the Q-functions graphically here.

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[1]) The global Fano factor gives, in general, only a prediction of the mean value of the time-dependent Fano factor, as the time development exhibits typically an oscillatory character. In the no-energy-transfer case, however, the Fano factors almost do not oscillate and the global Fano factor coincides with the quasi-steady state of the Fano factor.

The Husimi Q-functions are very wide, therefore no direct linearization of the quantum problem is possible and, probably, no pure quantum technique can yield the observed "magic" values $F_1 \approx 1.81$ and $F_3 \approx 0.81$. The only answer to our question concerning the quantitative sub-Poissonian behaviour can be obtained, somewhat surprisingly, by applying the method of classical trajectories as will be shown in the following.

3 Classical trajectory analysis

It is well known that the classical solution of THG is simpler than the quantum one and can serve as a good approximation for strong fields. It gives correct predictions of the output light intensities and frequency-conversion efficiency. Unfortunately, it cannot yield any information about the noise and statistical properties of the generated light, and the quantum model has to be adopted in the analysis. The method of classical trajectories [14,15] is of course based on the deterministic classical solutions but the quantum noise can be introduced artificially to the model and simulated by Gaussian distribution of initial amplitudes. For inputs strong in comparison to quantum noise, the method gives results in a very good agreement with the quantum-optical predictions as, e.g., graphically compared in Ref. 5.

In the classical trajectory approach, one assumes the input stochastic amplitudes of the fundamental mode in the form $\alpha_1 = \alpha_{10} + x_1 + \mathrm{i}y_1$ and of the third-harmonic mode as $\alpha_3 = \alpha_{30} + x_3 + \mathrm{i}y_3$, where x_k and y_k are real Gaussian stochastic quantities with the identical variances $\sigma^2 = 1/4$. With this choice of the noise variance, we find the expected values of quadrature squeezing $\mathrm{Var}(X_k) = \mathrm{Var}\left(\alpha_k e^{-\mathrm{i}\theta} + \alpha_k^* e^{\mathrm{i}\theta}\right) = 2(\overline{x_k^2} + \overline{y_k^2}) = 4\sigma^2 = 1$ and the Fano factor

$$F_k \equiv \frac{\mathrm{Var}\,(n_k)}{\overline{n_k}} = \frac{\overline{\alpha_k^{*2}\alpha_k^2} - \left(\overline{\alpha_k^*\alpha_k}\right)^2}{\overline{\alpha_k^*\alpha_k}} = \frac{4\sigma^2\left(\alpha_{k0}^2 + \sigma^2\right)}{\alpha_{k0}^2 + 2\sigma^2}$$
$$= 4\sigma^2\left(1 - \frac{\sigma^2}{\alpha_{k0}^2} + \cdots\right) \approx 4\sigma^2 = 1. \tag{4}$$

Here, we have also assumed that the field is strong in comparison to its noise, i.e., $\alpha_{k0}^2 \gg \sigma^2$. In this approach, one needs to solve numerically or analytically thousands of the classical THG trajectories. Then, the mean values are simply obtained by averaging (as denoted by the horizontal bar over the quantities) over all the trajectories.

Let us now recall the classical results of the third-harmonic generation process described by the following system of the two complex differential equations [9]

$$\begin{aligned} \dot{\alpha}_1 &= -3\mathrm{i}g\alpha_1^{*2}\alpha_3\,, \\ \dot{\alpha}_3 &= -\mathrm{i}g\alpha_1^3\,. \end{aligned} \tag{5}$$

By introducing real amplitudes and phases, $\alpha_k = r_k e^{\mathrm{i}\phi_k}$, Eq. (5) can be transformed into the system of the following three real equations

$$\begin{aligned} \dot{r}_1 &= -3gr_1^2 r_3 \sin\theta, \\ \dot{r}_3 &= gr_1^3 \sin\theta, \\ \dot{\theta} &= g\left(\frac{r_1^3}{r_3} - 9r_1 r_3\right)\cos\theta, \end{aligned} \tag{6}$$

where $\theta = 3\phi_1 - \phi_3$. The system of Eqs. (6) has two integrals of motion:

$$\begin{aligned} E &= r_1^2 + 3r_3^2 = n_1 + 3n_3\,, \\ \Gamma &= r_1^3 r_3 \cos\theta\,. \end{aligned} \tag{7}$$

By extracting r_1 and θ from (6), the equation for the amplitude r_3 is obtained as $(r_3\dot{r}_3/g)^2 + \Gamma^2 = r_3^2\left(E - 3r_3^2\right)^3$. Yet simpler form has the equation

$$\left(\frac{\dot{n}_3}{2g}\right)^2 + \Gamma^2 = n_3\left(E - 3n_3\right)^3 \tag{8}$$

for the intensity $n_3 = r_3^2$. Equation (8) can be solved by separation of variables as

$$2g\mathrm{d}t = \frac{\mathrm{d}n_3}{\sqrt{n_3\left(E - 3n_3\right)^3 - \Gamma^2}}\,. \tag{9}$$

The solutions are periodic and can be expressed by Jacobi elliptical functions. The general solutions have rather complicated and lengthy structure. But considering the purpose of our paper, it is sufficient to analyze their special cases only.

Two elementary solutions of Eq. (6) can be found easily. One of them is obtained for the initial phase mismatch $\theta = \frac{\pi}{2}$. Here, we find that $\Gamma = 0$, and

$$\begin{aligned} r_1(t) &= r\sqrt{\frac{1}{3\left(gr^2t\right)^2 + 1}}\,, \\ r_3(t) &= r\sqrt{\frac{\left(gr^2t\right)^2}{3\left(gr^2t\right)^2 + 1}}\,. \end{aligned} \tag{10}$$

This monotonic solution represents the third-harmonic generation from vacuum ($\alpha_3 = 0$), which corresponds to the well-known hyperbolic secant and tangent solution of SHG process. The second elementary solution is obtained for the initial zero phase mismatch $\theta = 0$ and the initial amplitudes satisfying the condition $r_1 = 3r_3$. The solution reads

$$\begin{aligned} \alpha_1(t) &= 3r\exp(-9\mathrm{i}gr^2t + \mathrm{i}\varphi), \\ \alpha_3(t) &= r\exp(-27\mathrm{i}gr^2t + 3\mathrm{i}\varphi). \end{aligned} \tag{11}$$

Eq. (11) represents the so-called *no-energy-transfer* solution, since the amplitude and energy in every interacting mode remain constant [13]. We apply the classical trajectory analysis for the *no-energy-transfer* solution (11). Thus, we assume the initial amplitudes $\alpha_1 = 3r$ and $\alpha_3 = r$ and blur them by Gaussian noise, which results in $\alpha_1 = 3r + x_1 + iy_1$ and $\alpha_3 = r + x_3 + iy_3$. The classical integrals of motion, given by Eq. (7), can be rearranged in the form of successive corrections

$$\begin{aligned} E &= 12r^2 + \Delta E_1 + \Delta E_0, \\ \Gamma &= 27r^4 + \Delta\Gamma_3 + \Delta\Gamma_2 + \Delta\Gamma_1 + \Delta\Gamma_0, \end{aligned} \tag{12}$$

where $\Delta E_1 = 6r\,(x_1 + x_3)$; $\Delta E_0 = x_1^2 + y_1^2 + 3\left(x_3^2 + y_3^2\right)$; $\Delta\Gamma_3 = 27r^3\,(x_1 + x_3)$, and $\Delta\Gamma_2 = 9r^2\left(x_1^2 - y_1^2 + 3x_1x_3 + 3y_1y_3\right)$. The lower-order terms $\Delta\Gamma_1$ and $\Delta\Gamma_0$ can be neglected in further considerations. The denominator in Eq. (9), after substitution of $n_3 = \frac{1}{12}E + \epsilon$ with some small correction ϵ, can be approximated by the quadratic function, $n_3\,(E - 3n_3)^3 - \Gamma^2 \approx \frac{9}{256}E^4 - \frac{27}{8}E^2\epsilon^2 - \Gamma^2 = \frac{27}{8}E^2\left(A^2 - \epsilon^2\right)$ if one neglects the higher-order terms involving ϵ^3 and ϵ^4. Under this approximation, we can integrate Eq. (9), which finally leads to the simple result

$$\begin{aligned} n_3\,(t) &= \frac{E}{12} + A\cos\left(\Omega g t\right) \\ &= r^2 + B + A\cos\left(\Omega g t\right), \end{aligned} \tag{13}$$

where $\Omega = \sqrt{\frac{27}{2}E}$, $A = \frac{1}{2}r\sqrt{6\,(y_3 - y_1)^2 + (x_1 - 3x_3)^2}$, and $B = \frac{1}{12}\Delta E_1 = \frac{1}{2}r\,(x_1 + x_3)$. For the fundamental (or subharmonic) mode we obtain the similar result

$$n_1\,(t) = E - 3n_3 = 9r^2 + 9B - 3A\cos\left(\Omega g t\right). \tag{14}$$

Both solutions (13) and (14) are given by constants weakly perturbed by a harmonic function. Due to the frequency dispersion, the different solutions are drifting in phase and create a banana-shaped cloud in the phase space, which for longer evolution times goes over into a full ring [5]. Now, one can perform the averaging of the solutions (13) and (14) to calculate the requested moments. We get

$$\begin{aligned} \overline{n_1} &= 9r^2, \quad \overline{n_1^2} = 81r^4 + 81\overline{B^2} + \tfrac{9}{2}\overline{A^2}, \\ \overline{n_3} &= r^2, \quad \overline{n_3^2} = r^4 + \overline{B^2} + \tfrac{1}{2}\overline{A^2}. \end{aligned} \tag{15}$$

where $\overline{A^2} = \frac{11}{2}r^2\sigma^2 = \frac{11}{8}r^2$ and $\overline{B^2} = \frac{1}{2}r^2\sigma^2 = \frac{1}{8}r^2$, whereas $\overline{B} = 0$ and $\overline{\cos^2\left(\Omega g t\right)} = \frac{1}{2}$. Finally, we arrive at the Fano factors in the form of rational numbers

$$\begin{aligned} F_1 &= \frac{1}{r^2}\left(9\overline{B^2} + \frac{1}{2}\overline{A^2}\right) = \frac{9}{8} + \frac{1}{2}\frac{11}{8} = \frac{29}{16}, \\ F_3 &= \frac{1}{r^2}\left(\overline{B^2} + \frac{1}{2}\overline{A^2}\right) = \frac{1}{8} + \frac{1}{2}\frac{11}{8} = \frac{13}{16}. \end{aligned} \tag{16}$$

These numbers agree very well with the values obtained by numerical study of the quantum problem.

4 Conclusions

We have shown numerically that, for a special choice of initial states $\alpha_1 = 3r, \alpha_3 = r, r > 1$ and the phase mismatch $\theta \simeq 0$, the third-harmonic mode exhibits sub-Poissonian photon-number statistics with the Fano factor $F_3 \simeq 0.81$. This value remains unchanged for sufficiently strong input amplitudes $r > 1$ during the whole time evolution, $gt \gtrsim 1$. Similarly, the Fano factor of the fundamental mode also exhibits relatively small and stable noise, but is super-Poissonian with $F_1 \simeq 1.81$. We have shown that these results can also be derived by the method of classical trajectories in the approximation of strong field. This method gives the results $F_1 = \frac{29}{16} = 1.8125$, $F_3 = \frac{13}{16} = 0.8125$ surprisingly similar to those obtained in the quantum model by solving the Schrödinger equation.

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