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Noise, instability and squeezing in third harmonic generation

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Abstract

We present a quantum theory of intracavity third harmonic generation. Third-order noise is included into the consideration and the corresponding Langevin equations of motion for the stochastic amplitudes of the interacting modes are derived. Semiclassical steady-state solutions are found, linear stability analysis is carried out, and self-pulsing temporal behavior of the fundamental and third harmonic modes is found in the instability domain beyond the critical point. Quadrature amplitude squeezing spectra are calculated, and it is shown that perfect squeezing is approached in both the fundamental and the third harmonic modes in the vicinity of the critical point. Higher-order moments of the field operators and manifestations of the third-order noise in photon correlation phenomena are also discussed.

1. Introduction

Generation of third harmonics in crystals or atomic gases placed inside a cavity represents an interesting example for investigating quantum noise and instability in nonlinear optical processes. It is known that a consistent quantum theory of nonlinear interaction of radiation field modes in the cavity is based on the Fokker–Planck equation for a quasi-probability distribution or on equivalent Langevin equations for stochastic amplitudes. For a number of processes, such as second harmonic generation, parametric oscillation, four-wave mixing and several others, the mentioned approach turned to be very effective due to utilization of the concept of second-order noise sources. For the latter ones only the two-time correlators, which are proportional to a δ-function in the case of white noise, do not vanish (see, for example, Ref. [1]). However, for higher order nonlinear processes it is necessary to deal with the concept of higher-order noise, which is not well studied in quantum optics.

In the present work we are going to examine this problem for the process of third harmonic generation (THG) in a $\chi^{(3)}$ nonlinear medium placed in a cavity. Another purpose of the work is to investigate the squeezing and the temporal instability effects in the THG. Note that both these effects are well known for the process of second harmonic generation (SHG). The instability in SHG and self-pulsing behavior of the intensities of the fundamental and second harmonic modes have been predicted in Ref. [2]. Second harmonic generation was also one of the first processes which were considered for the production of squeezed light. Among the recent works we note Refs. [3,4], where 52% squeezing in the fundamental mode and 30% squeezing in the second harmonic mode in a doubly and singly resonant cavity

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configuration, respectively, have been obtained. Although it is obvious that the experiments for the THG are considerably harder, due to low $\chi^{(3)}$ nonlinearities and phase matching difficulties, however, recent results [5,6] indicate definite progress and increasing interest in this subject.

2. Basic equations; third-order noise

We consider a doubly resonant THG in which three photons of frequency ω_1 in the fundamental mode a_1 can annihilate to produce a photon of frequency $\omega_2 = 3\omega_1$ in the third harmonic mode a_2. The fundamental mode is resonantly driven by an external classical field. The interaction of the fundamental mode with that of the third harmonic in a nonlinear $\chi^{(3)}$ medium is described by the following Hamiltonian:

$$
\begin{aligned}
H &= \hbar\omega_1 a_1^+ a_1 + 3\hbar\omega_1 a_2^+ a_2 + H_{\rm int} + H_{\rm loss}, \\
H_{\rm int} &= \mathrm{i}\hbar\chi\left(a_1^3 a_2^+ - a_1^{+3} a_2\right) \\
&\quad + \mathrm{i}\hbar\left(E\mathrm{e}^{-\mathrm{i}\omega_1 t} a_1^+ - E^*\mathrm{e}^{\mathrm{i}\omega_1 t} a_1\right), \\
H_{\rm loss} &= a_1\Gamma_1^+ + a_1^+\Gamma_1 + a_2\Gamma_2^+ + a_2^+\Gamma_2,
\end{aligned} \tag{1}
$$

where χ is the resulting coupling constant proportional to the third-order susceptibility $\chi^{(3)}$, E is the amplitude of the driving field at the frequency ω_1, and Γ_i, Γ_i^+ $(i = 1, 2)$ are reservoir operators for the fundamental and third harmonic modes, which will give rise to the cavity damping constants γ_1 and γ_2, respectively.

Using the standard technique, we first eliminate adiabatically the reservoir operators and derive the master equation for the density operator ρ of two modes of the radiation field in the interaction picture. This equation is then transformed into a Fokker–Planck equation for the quasi-probability distribution function $P(\boldsymbol{x})$ in a generalized P-representation [1,7], which we write in the general form introducing a four-component variable $\boldsymbol{x} \equiv (x_1, x_2, x_3, x_4) \equiv (\alpha_1, \beta_1, \alpha_2, \beta_2)$:

$$
\frac{\partial P(\boldsymbol{x})}{\partial t} = \left(-\sum_i \frac{\partial}{\partial x_i} A_i(\boldsymbol{x}) + \frac{1}{2}\sum_{i,j} \frac{\partial^2}{\partial x_i \partial x_j} D_{i,j}(\boldsymbol{x}) - \frac{1}{6}\sum_{i,j,k} \frac{\partial^3}{\partial x_i \partial x_j \partial x_k} C_{ijk}(\boldsymbol{x}) \right) P(\boldsymbol{x}). \tag{2}
$$

Quantities α_1, β_1, α_2 and β_2 are independent complex variables corresponding to the operators a_1, a_1^+, a_2 and a_2^+, respectively. The elements of the drift vector are equal to

$$
\begin{aligned}
A_1 &= E - \gamma_1\alpha_1 - 3\chi\beta_1^2\alpha_2, \\
A_2 &= E^* - \gamma_1\beta_1 - 3\chi\alpha_1^2\beta_2, \\
A_3 &= -\gamma_2\alpha_2 + \chi\alpha_1^3, \quad A_4 = -\gamma_2\beta_2 + \chi\beta_1^3,
\end{aligned} \tag{3}
$$

and the nonvanishing noise terms are

$$
D_{11} = -6\chi\beta_1\alpha_2, \qquad D_{22} = -6\chi\alpha_1\beta_2, \tag{4}
$$

$$
C_{111} = -6\chi\alpha_2, \qquad C_{222} = -6\chi\beta_2. \tag{5}
$$

Eq. (2) has a complicated form due to the third-order derivatives. We can obtain stochastic equations of motion which are equivalent to Eq. (2), following the method of Ito (see, for example Ref. [1]). This method, when applied to Eq. (2), is not so simple and well known as in the case of diffusion equations without the tensor C_{ijk}, and it leads naturally to the concept of third-order noise sources. For the one-dimensional problem, the stochastic equation approach containing third-order noise has been developed by Gardiner [1]. For the general multi-dimensional case of Eq. (2), it can be shown that the system of equivalent stochastic equations for x_i-variables has the following form:

$$
\frac{\partial x_i}{\partial t} = A_i(\boldsymbol{x}) + \sum_j B_{ij}(\boldsymbol{x})\xi_j(t) + \sum_{n,m} R_{inm}(\boldsymbol{x})\eta_{nm}(t), \tag{6}
$$

where the noise terms $\xi_j(t)$ and $\eta_{nm}(t)$ correspond to the contributions, arising from the second- and third-order derivatives, respectively, in the Fokker–Planck equation (2). The nonvanishing correlation functions for them are

$$
\begin{aligned}
&\langle \xi_i(t)\xi_j(t')\rangle = \delta_{ij}\delta(t-t'), \\
&\langle \eta_{im}(t)\eta_{jn}(t')\eta_{kl}(t'')\rangle \\
&\quad = \delta_{ij}\delta_{jk}\delta_{mn}\delta_{nl}\delta(t-t')\delta(t'-t'').
\end{aligned} \tag{7}
$$

Coefficients B_{ij} and D_{imn} of the system of equations (6) are obtained from the relations

$$
\sum_k B_{ik}B_{jk} = D_{ij}, \qquad \sum_{n,m} R_{inm}R_{jnm}R_{knm} = C_{ijk}. \tag{8}
$$

Eqs. (2), (6) and (7), (8) are written in a general form. For the case of THG Eqs. (3)–(5) with account of Eq. (8) give

$$B_{11}=\sqrt{D_{11}},\qquad B_{22}=\sqrt{D_{22}},$$
$$R_{111}=\sqrt[3]{C_{111}},\qquad R_{222}=\sqrt[3]{C_{222}} \tag{9}$$

and lead to the following Langevin equations:

$$\frac{\partial\alpha_1}{\partial t}=A_1+\sqrt{-6\chi\beta_1\alpha_2}\,\xi_1(t)+\sqrt[3]{-6\chi\alpha_2}\,\eta_{11}(t),$$
$$\frac{\partial\beta_1}{\partial t}=A_2+\sqrt{-6\chi\alpha_1\beta_2}\,\xi_2(t)+\sqrt[3]{-6\chi\beta_2}\,\eta_{22}(t),$$
$$\frac{\partial\alpha_2}{\partial t}=A_3,$$
$$\frac{\partial\beta_2}{\partial t}=A_4. \tag{10}$$

3. Semiclassical steady states and instability

To analyze the system of equations (10) it is convenient to transform them to the photon number and phase variables of the modes:

$$n_j=\alpha_j\beta_j,\qquad \varphi_j=\frac{1}{2\mathrm{i}}\ln\left(\frac{\alpha_j}{\beta_j}\right)\qquad (j=1,2). \tag{11}$$

It can be shown that the variable change formula (Ito form) in the case of stochastic equations with third order noise terms is

$$\frac{\mathrm{d}f(x)}{\mathrm{d}t}=\sum_i\frac{\partial f}{\partial x_i}A_i(x)+\frac{1}{2}\sum_{i,j}\frac{\partial^2 f}{\partial x_i\partial x_j}D_{ij}(x)$$
$$+\frac{1}{6}\sum_{i,j,k}\frac{\partial^3 f}{\partial x_i\partial x_j\partial x_k}C_{ijk}(x)$$
$$+\sum_i\frac{\partial f}{\partial x_i}\left(\sum_k B_{ik}(x)\xi_k(t)+\sum_{n,m}R_{inm}(x)\eta_{nm}(t)\right). \tag{12}$$

In the variables (11) the equations of motion become

$$\frac{\partial n_1}{\partial t}=2|E|n_1^{1/2}\cos(\varphi-\varphi_1)-2\gamma_1 n_1$$
$$-6\chi n_1^{3/2}n_2^{1/2}\cos(3\varphi_1-\varphi_2)+\Gamma(t),$$
$$\frac{\partial n_2}{\partial t}=-2\gamma_2 n_2+2\chi n_1^{3/2}n_2^{1/2}\cos(3\varphi_1-\varphi_2),$$
$$\frac{\partial\varphi_1}{\partial t}=\frac{|E|}{\sqrt{n_1}}\sin(\varphi-\varphi_1)$$
$$+\left(3\chi n_1^{1/2}n_2^{1/2}-3\chi n_1^{-1/2}n_2^{1/2}\right.$$
$$\left.+2\chi n_1^{-3/2}n_2^{1/2}\right)\sin(3\varphi_1-\varphi_2)+\Phi(t),$$
$$\frac{\partial\varphi_2}{\partial t}=\chi n_1^{3/2}n_2^{-1/2}\sin(3\varphi_1-\varphi_2), \tag{13}$$

where φ is the phase of the driving field $E=|E|\exp(\mathrm{i}\varphi)$, and the noise terms are

$$\Gamma(t)=\beta_1 B_{11}\xi_1(t)+\alpha_1 B_{22}\xi_2(t)$$
$$+\beta_1 R_{111}\eta_{11}(t)+\alpha_1 R_{222}\eta_{22}(t),$$
$$\Phi(t)=\frac{1}{2\mathrm{i}}\left(\frac{B_{11}}{\alpha_1}\xi_1(t)-\frac{B_{22}}{\beta_1}\xi_2(t)\right.$$
$$\left.+\frac{R_{111}}{\alpha_1}\eta_{11}(t)-\frac{R_{222}}{\beta_1}\eta_{22}(t)\right). \tag{14}$$

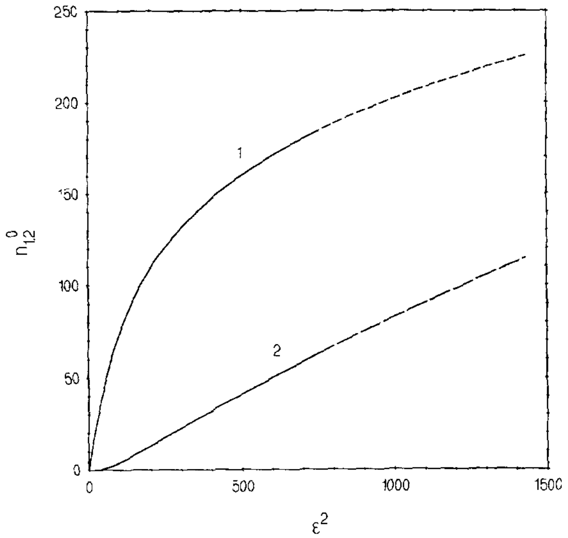

Fig. 1. Semiclassical steady-state photon numbers for the fundamental and third harmonic modes n_1^0 (curve 1) and n_2^0 (curve 2) versus the pump intensity parameter ε^2 for $k=10^{-5}$ and $r=1$. The solid parts of the curves correspond to the stable solutions, while the broken parts describe the unstable region.

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Analysis of the steady-state solutions n_j^0 and φ_j^0 $(j=1,2)$ of these equations in the semiclassical approximation and of conditions of their stability with respect to small fluctuations (linear stability analysis) leads to the following results:

$$\varphi_1^0=\varphi_2^0/3=\varphi,\qquad 3k\left(n_1^0\right)^{5/2}+\left(n_1^0\right)^{1/2}=\varepsilon,$$
$$n_2^0=\frac{k}{r}\left(n_1^0\right)^3, \tag{15}$$

where we have used the following notations:

$$r=\gamma_2/\gamma_1,\qquad k=\chi^2/\gamma_1\gamma_2,\qquad \varepsilon=|E|/\gamma_1. \tag{16}$$

This solution is stable if $\varepsilon<\varepsilon_c$, where

$$\varepsilon_c=\frac{1}{\sqrt[4]{6k}}\left[(1+r)^{1/4}+\tfrac{1}{2}(1+r)^{5/4}\right] \tag{17}$$

is the critical point. The critical photon number in the fundamental mode is

$$n_1^c=\left(\frac{1+r}{6k}\right)^{1/2}, \tag{18}$$

and the dependence of n_1^0 and n_2^0 on $|\varepsilon|^2$ is shown in Fig. 1. Beyond the critical point ($\varepsilon>\varepsilon_c$), i.e. for sufficiently strong driving field, the real parts of the eigenvalues of the matrices of the linearized equations of motion (see Eqs. (19), (20)) become negative leading to instability. In the instability region the intensities of the third harmonic and the fundamental modes exhibit self-pulsing temporal behavior. The mode dynamics depends on the initial conditions and is illustrated in Fig. 2 for a particular realization.

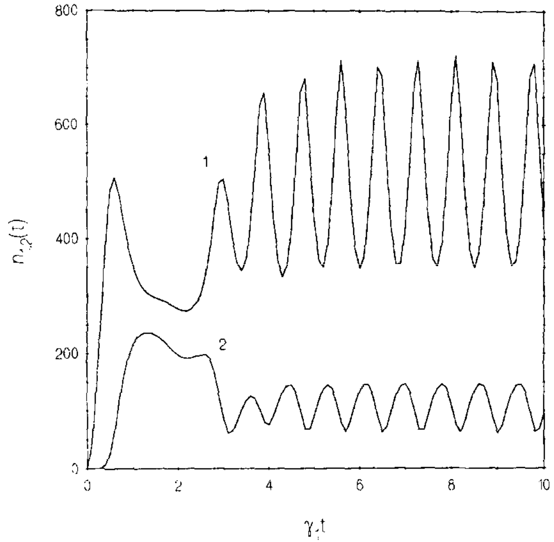

Fig. 2. Demonstration of self-pulsing behavior of the fundamental and third harmonic photon numbers $n_1(t)$ (curve 1) and $n_2(t)$ (curve 2) versus $\gamma_1 t$: numerical solution of Eqs. (10) in the semiclassical approximation for $k=10^{-5}$, $r=1$, $\varepsilon=60$ and initial conditions $\alpha_1(0)=1+i$, $\alpha_2(0)=0$.

4. Linearized treatment of quantum fluctuations and squeezing

The linearized equations for small deviations $\delta n_j=n_j-n_j^0$, $\delta\varphi_j=\varphi_j-\varphi_j^0$ from the steady states, which are applicable in the region $\varepsilon<\varepsilon_c$ and under the condition $n_j^0\gg1$, are

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix}\delta n_1\\ \delta n_2\end{pmatrix}=-A_n\begin{pmatrix}\delta n_1\\ \delta n_2\end{pmatrix}+\begin{pmatrix}\Gamma^0\\ 0\end{pmatrix}, \tag{19}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix}\delta\varphi_1\\ \delta\varphi_2\end{pmatrix}=-A_\varphi\begin{pmatrix}\delta\varphi_1\\ \delta\varphi_2\end{pmatrix}+\begin{pmatrix}\Phi^0\\ 0\end{pmatrix}, \tag{20}$$

where the matrices A_n and A_φ are

$$A_n=\begin{pmatrix}\gamma_1+\dfrac{6\chi^2}{\gamma_2}\left(n_1^0\right)^2 & 3\gamma_2\\ -\dfrac{3\chi^2}{\gamma_2}\left(n_1^0\right)^2 & \gamma_2\end{pmatrix},$$

$$A_\varphi=\begin{pmatrix}\gamma_1-\dfrac{6\chi^2}{\gamma_2}\left(n_1^0\right)^2 & \dfrac{3\chi^2}{\gamma_2}\left(n_1^0\right)^2\\ -3\gamma_2 & \gamma_2\end{pmatrix}. \tag{21}$$

The nonzero noise correlators are obtained with use of the Eqs. (14), (7) to be equal to

$$\langle\Gamma^0(t)\Gamma^0(t')\rangle=-12\frac{\chi^2}{\gamma_2}\left(n_1^0\right)^3\delta(t-t'),$$

$$\langle\Phi^0(t)\Phi^0(t')\rangle=\frac{3\chi^2}{\gamma_2}n_1^0\delta(t-t'),$$

$$\langle\Gamma^0(t)\Gamma^0(t')\Gamma^0(t'')\rangle$$
$$=-12\frac{\chi^2}{\gamma_2}\left(n_1^0\right)^3\delta(t-t')\delta(t'-t''),$$

$$\langle\Gamma^0(t)\Phi^0(t')\Phi^0(t'')\rangle$$
$$=\frac{3\chi^2}{\gamma_2}n_1^0\delta(t-t')\delta(t'-t''). \tag{22}$$

Let us now turn to the problem of squeezing of quantum fluctuations in the quadrature phase amplitudes $X_i^{\vartheta} = a_i \exp(-i\vartheta_i) + a_i^{+} \exp(i\vartheta_i)$ of the fundamental and the third harmonic modes $(i = 1, 2)$. We calculate the corresponding squeezing spectra $S_1(\omega)$ and $S_2(\omega)$ using the standard approach (see, for example, Ref. [8]). As our calculations show, the squeezing effect is maximal for the amplitude fluctuations, i.e. when the squeezing spectra $S_i(\omega)$ are expressed in terms of the photon number fluctuations and turn out to be equal to

$$S_1(\omega) = 1 + \frac{2\gamma_1}{n_1^0}\langle \delta n_1(-\omega)\delta n_1(\omega)\rangle = 1 - \frac{24k(n_1^0)^2\left[r^2 + (\omega/\gamma_1)^2\right]}{d_1(\omega)},$$

$$S_2(\omega) = 1 + \frac{2\gamma_2}{n_2^0}\langle \delta n_2(-\omega)\delta n_2(\omega)\rangle = 1 - \frac{216k^2(n_1^0)^4}{d_2(\omega)}, \qquad (23)$$

where $\delta n_i(\omega)$ are Fourier components of the photon number fluctuations $\delta n_i(t)$, and the quantities $d_{1,2}(\omega)$ are

$$d_1(\omega) = \left[r + 15kr(n_1^0)^2 - (\omega/\gamma_1)^2\right]^2 + \left[1 + r + 6k(n_1^0)^2\right]^2(\omega/\gamma_1)^2,$$

$$d_2(\omega) = \left[1 + 15k(n_1^0)^2 - r(\omega/\gamma_2)^2\right]^2 + \left[1 + r + 6k(n_1^0)^2\right]^2(\omega/\gamma_2)^2. \qquad (24)$$

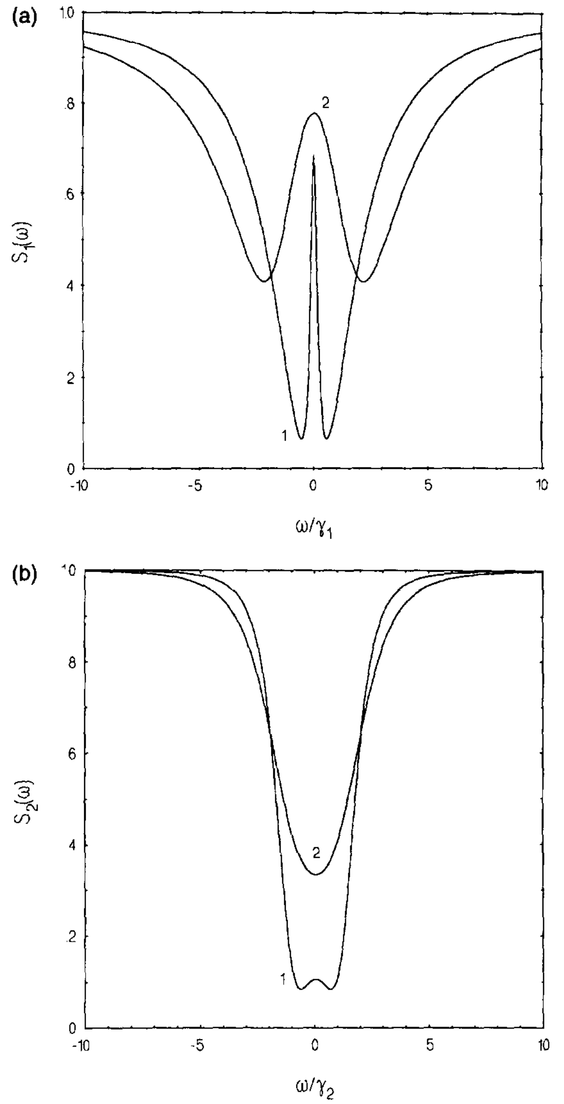

Fig. 3. Amplitude squeezing spectra $S_i(\omega)$ versus ω/γ_i for the fundamental (a) and third harmonic modes (b), respectively, at the critical point. (a) $r = 0.1$ (1), $r = 1$ (2). (b) $r = 10$ (1), $r = 1$ (2).

The maximal reduction of quantum fluctuations below the shot-noise level $(S_i(\omega) < 1)$ is achieved in the vicinity of the critical point, in accordance with a number of works dealing with other nonlinear optical processes (see, for example, Ref. [8]). If $r \ll 1$, perfect squeezing is approached in the fundamental mode at the sideband frequencies, which are, however, very close to the zero frequency $(\omega/\gamma_1 \cong 0)$ in the limit $r \to 0$. In contrast, if $r \gg 1$ the perfect squeezing is approached in the third harmonic mode at the sideband frequencies $\omega \cong \pm\gamma_2/\sqrt{2}$. For the values of parameters represented in Fig. 3 by curves 1, we see that the noise reduction effect exceeds 90%. We note that the third-order quantum noise does not contribute to the squeezing spectra (23) in the lowest order with respect to small fluctuations.

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5. Higher-order moments and third-order noise in photon correlations

This section concerns with a discussion of the role of the third order in physical quantities. As it may be expected, the third-order noise effects will be manifested in higher-order operator moments, which describe, in particular, the photon correlation phenomena.

At first let us consider this question in the small time approximation $t \ll \gamma_{1,2}^{-1}$. For this purpose we turn to the solution of stochastic equations of motion (10) for small time intervals in the case of vacuum initial conditions $\alpha_i(t=0) = \beta_i(t=0) = 0$ $(i = 1, 2)$. Writing down the amplitudes of the fundamental mode in the form

$$\alpha_1(t) = \alpha_1^{\mathrm{d}}(t) + F_1(t),$$
$$\beta_1(t) = \beta_1^{\mathrm{d}}(t) + F_2(t), \tag{25}$$

where we separate the noise contributions $F_{1,2}$ from the deterministic parts α_1^{d} and β_1^{d}, we use then the Euler iteration method. This yields, in the third order of iteration:

$$\alpha_1^{\mathrm{d}}(\tau) = \beta_1^{\mathrm{d}}(\tau) = \varepsilon\tau - \frac{\varepsilon\tau^2}{3} + \varepsilon\left(\frac{\tau}{3}\right)^3 - 4kr\varepsilon^5\frac{\tau^7}{3^6} + \dots, \tag{26}$$

$$F_i(\tau) = \sqrt[3]{-6kr}\,\varepsilon W_i(\tau)\left(\frac{\tau}{3}\right)^{5/3} + \sqrt{-12kr}\,\varepsilon^2 V_i(\tau)\left(\frac{\tau}{3}\right)^3, \tag{27}$$

where $\tau = \gamma_1 t$ and we have chosen $\varepsilon^* = \varepsilon$ for simplicity. $V_i(\tau)$ and $W_i(\tau)$ are the noise sources of the second and the third order, respectively, which are connected with $\xi_i(t)$ and $\eta_{ii}(t)$ by

$$\xi_i(t)\,\mathrm{d}t = V_i(t)\sqrt{\mathrm{d}t}, \qquad \eta_{ii}(t)\,\mathrm{d}t = W_i(t)\sqrt[3]{\mathrm{d}t}. \tag{28}$$

Their nonzero correlators are

$$\langle V_i(t)^2\rangle = \langle W_i(t)^3\rangle = 1. \tag{29}$$

Using the results (26)–(29) we calculate the second- and the third-order correlators for the fundamental mode. The normalized second-order correlator

$$g_1^{(2)} = \frac{\langle (a_1^+)^2 a_1^2\rangle}{\langle a_1^+ a_1\rangle^2}, \tag{30}$$

in the lowest approximation in τ contains a contribution from the second-order noise source and is determined by

$$g_1^{(2)} - 1 \approx \frac{2\langle F_1^2\rangle}{\left(\alpha_1^{\mathrm{d}}\right)^2} \approx -\frac{8}{3}kr\varepsilon^2\left(\frac{\tau}{3}\right)^4 + \dots. \tag{31}$$

We see that the quantity $g_1^{(2)} - 1$ is proportional to the squared amplitude of the driving field and that the pair correlation of photons is of antibunched character.

As to the normalized third-order correlator

$$g_1^{(3)} = \frac{\langle (a_1^+)^3 a_1^3\rangle}{\langle a_1^+ a_1\rangle^3}, \tag{32}$$

the leading contribution, in the lowest order in τ, arises from the third-order noise source. Indeed, with use of the solutions (26), (27), we obtain

$$g_1^{(3)} - 1 \approx \frac{1}{\left(\alpha_1^{\mathrm{d}}\right)^3}\left(\langle F_1^3\rangle + 3\alpha_1^{\mathrm{d}}\langle F_1^2\rangle\right) \approx -\frac{4}{9}kr\left(\frac{\tau}{3}\right)^2 - 8kr\varepsilon^2\left(\frac{\tau}{3}\right)^4 + \dots, \tag{33}$$

where the first term is originated from the third-order noise source W_1, while the second one is determined by the second-order noise source V_1. Note that the correlation of photons, described by $g_1^{(3)}$, is also of antibunched character.

For the case of the third-harmonic mode one may arrive at similar conclusions on the role of the third-order noise, but the corresponding results for $g_2^{(2)}$ and $g_2^{(3)}$ are more complicated to be reproduced here.

For completeness of our discussion, we turn also to the results, containing the effects of the third-order noise, in the steady-state regime. We calculate, in particular, the so-called coefficient of asymmetry, which is determined, in the lowest order of linear

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approximation of quantum fluctuations, by the following expression:

$$\langle (a_i^+ a_i - \langle a_i^+ a_i \rangle)^3 \rangle = n_i^0 \left[1 + \frac{3\langle \delta n_i^2 \rangle}{n_i^0} + \frac{\langle \delta n_i^3 \rangle}{n_i^0} \right]. \tag{34}$$

The quantities $\langle \delta n_i^2 \rangle$ and $\langle \delta n_i^3 \rangle$ may be calculated with use of the solution of the linearized equations of motion (19) in the steady-state limit $t \gg \gamma_{1,2}^{-1}$:

$$\begin{pmatrix} \delta n_1(t) \\ \delta n_2(t) \end{pmatrix} = \int_{-\infty}^{t} \mathrm{d}t' \sum_{k=1}^{2} \mathrm{e}^{-\lambda_k (t-t')} \frac{(A_n - \lambda_{3-k} I)}{\lambda_k - \lambda_{3-k}} \times \begin{pmatrix} \Gamma_0(t') \\ 0 \end{pmatrix}. \tag{35}$$

Here the matrix A_n is determined by Eq. (21), I is the identity matrix, and $\lambda_{1,2}$ are the eigenvalues of A_n.

To illustrate our results we give here relatively simple expressions obtained for the third harmonic mode:

$$\frac{\langle \delta n_2^2 \rangle}{n_2^0} = -12\gamma_2 \frac{[(A_n)_{21}]^2}{2\lambda_1 \lambda_2 (\lambda_1 + \lambda_2)}, \tag{36}$$

$$\frac{\langle \delta n_2^3 \rangle}{n_2^0} = -12\gamma_2 \frac{[(A_n)_{21}]^3}{(\lambda_1 - \lambda_2)^2} \times \left\{ \frac{3}{(2\lambda_1 + \lambda_2)(2\lambda_2 + \lambda_1)} - \frac{1}{3\lambda_1 \lambda_2} \right\}. \tag{37}$$

Direct numerical analysis of these results indicates that the relation $\langle \delta n_2^3 \rangle / \langle \delta n_2^2 \rangle$, evaluated, for example, in the neighborhood of the critical point, is of order 0.05. Hence, the third term in Eq. (34) may not be neglected if we set on the precision smaller than the magnitude of this relation. We note also that the second- and third-order correlators of the noise term $\Gamma_0(t')$ (see Eq. (22)), which have been used to calculate $\langle \delta n_2^2 \rangle$ and $\langle \delta n_2^3 \rangle$, respectively, are of the same magnitude. As a consequence, the small values of the ratio $\langle \delta n_2^3 \rangle / \langle \delta n_2^2 \rangle$ originate from the kinematics of the coefficients in Eq. (35).

In conclusion, we note that the third-order noise (along with the second-order one) is expected to play an essential role in the instability domain of the THG, as well as in the exact nonlinear treatment of quantum fluctuations. The research on this subject is in progress now.

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