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Exact quantum treatment of the parametric $\chi^{(3)}$-interaction

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Abstract. We present an exact quantum treatment of intracavity parametric four-wave mixing accompanied by self- and cross-phase modulation effects. A steady-state solution of the Fokker–Planck equation in a generalized P-representation is found for a generated signal mode. A general expression for arbitrary moments of the signal field operators is obtained and qualitative effects caused by non-linear treatment of quantum fluctuations are discussed. The behaviour of the exact steady-state intensity, as well as of the second-order correlation function and of the photon number fluctuations of the signal mode, is studied. A sub-Poissonian photon statistic is found in the above-threshold region and a critical increase in the photon number fluctuations is shown in the transition region for the case of relatively small non-linearities. Characteristic properties of the threshold behaviour are discussed in the case of large non-linearities.

1. Introduction

Current research on non-linear phenomena in quantum optics is known to be of fundamental and practical importance. Quantum optical non-linear systems are usually described within the framework of stochastic equations of motion for the field amplitudes with the use of a linearization procedure about the stable steady-state classical solutions. Such an approach was applied to studies of basic non-linear optical processes including intracavity four-wave mixing [1, 2], sub- and second-harmonic generation [1, 3–6], and optical bistability [7] (see also [8–11] and references therein). It made it possible to find in a semiclassical approximation the amplitudes of generated modes of a radiation field, to carry out a stability analysis and to find the operating regimes, as well as to study the effects of quantum fluctuations in the lowest approximation. However, the linearized theories have a limited range of applications: in particular, they are not valid in the regions of critical (threshold, turning, etc) points, where the quantum fluctuations increase dramatically.

An exact treatment of quantum fluctuations can be achieved via a solution of the Fokker-Planck equation for quasiprobability distribution functions. This approach gives the possibility of refining the results of linearized theories quantitatively, as well as predicting new qualitative phenomena for several non-linear optical systems [4, 7, 12]. However, the derivation of a quasiprobability distribution function is a difficult problem, which is actually equivalent to derivation of the density operator for the system.

In this paper we study an intracavity parametric interaction in a $\chi^{(3)}$-medium, and we show that non-linear treatment of the quantum fluctuations is successful. We present

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an exact quantum theory for the realistic model of four-wave mixing, which incorporates the effects of self-phase modulation and cross-phase modulation. These phase modulation effects would normally arise in a $\chi^{(3)}$-medium. However, they are usually neglected. Our aim is to find an exact steady-state solution of the Fokker–Planck equation for a generalized P-representation. Using this solution we calculate the operator moments for the generated signal field and study the behaviour of the signal field intensity, as well as of the second-order correlation function and of the photon number fluctuations.

The paper is organized as follows. In section 2 the model Hamiltonian is written and some of the previously obtained [13] semiclassical results are presented for convenience and for further comparison. In section 3 an exact steady-state solution of the Fokker–Planck equation in a generalized P-representation is derived and a general expression is obtained for arbitrary moments of the signal-mode amplitude operators. In sections 4 and 5 the results of calculation of the signal-mode intensity and of the second-order correlation function are presented and discussed.

2. The model Hamiltonian and semiclassical results

The system under consideration consists of a $\chi^{(3)}$-medium placed in a single-mode ring cavity with resonant frequency ω_c and damping constant γ. The non-linear medium is pumped by two copropagating monochromatic light beams of different frequencies ω_1 and ω_2. As a result of four-wave mixing (FWM) parametric interaction the pump fields excite an intracavity signal mode ω_c. The exact phase matching consideration determines a frequency $\omega_0 = \frac{1}{2}(\omega_1 + \omega_2)(\omega_0 \simeq \omega_c)$, which will appear (see section 3) as a rotating frame frequency for the signal field. In the general case we allow for a cavity detuning $\Delta = \omega_0 - \omega_c$. The ω_1 and ω_2 frequencies are considered to be located away from the cavity resonance, i.e. the cavity is transparent for the pump beams, which makes the single-pass effects in the pumps irrelevant. In this case we can neglect the pump depletion and treat the complex amplitudes E_1 and E_2 of the pump fields as fixed classical constants.

In addition to the parametric FWM we take into account the phase modulation effects. This is achieved naturally by consideration of the full $\chi^{(3)}$-interaction Hamiltonian:

$$H_{\rm int} \propto \chi^{(3)}(a^+ e^{i\omega_c t} + E_1^* e^{i\omega_1 t} + E_2^* e^{i\omega_2 t})^2 (a e^{-i\omega_c t} + E_1 e^{-i\omega_1 t} + E_2 e^{-\omega_2 t})^2 \tag{1}$$

where a^+ and a are creation and annihilation operators for the signal mode ω_c. Taking into account the phase-matching condition $\omega_1 + \omega_2 \simeq 2\omega_c$ and employing the rotating-wave approximation, we then neglect in (1) all fast oscillating terms, namely the terms containing exponents of the type $\exp(\pm i(\omega_c - \omega_{1,2}))$ and $\exp(\pm i(\omega_1 - \omega_2))$. This implies in fact that we assume a fulfillment of the following conditions: $|\omega_c - \omega_{1,2}| \gg |\omega_0 - \omega_c|$, $|\omega_1 - \omega_2| \gg |\omega_0 - \omega_c|$. Thus the interaction Hamiltonian, which governs the evolution of the a^+ and a operators, becomes

$$H_{\rm int} = \tfrac{1}{2}\hbar\chi(a^2 E_1^* E_2^* e^{i2\Delta t} + a^{+2} E_1 E_2 e^{-i2\Delta t}) + \tfrac{1}{4}\hbar\chi a^{+2} a^2 + \hbar\chi(|E_1|^2 + |E_2|^2) a^+ a \tag{2}$$

where χ is the resulting coupling constant, proportional to the third-order susceptibility $\chi^{(3)}$, and $\Delta = \omega_0 - \omega_c$ is the cavity detuning. The first term in (2) is responsible for the parametric FWM coupling, while the second and the third terms describe processes of self-phase modulation and cross-phase modulation, respectively.

Accounting for decay of the cavity mode we write the full Hamiltonian of the system as follows

$$H = \hbar\omega_c a^+ a + H_{\rm int} + a^+\Gamma + a\Gamma^+. \tag{3}$$

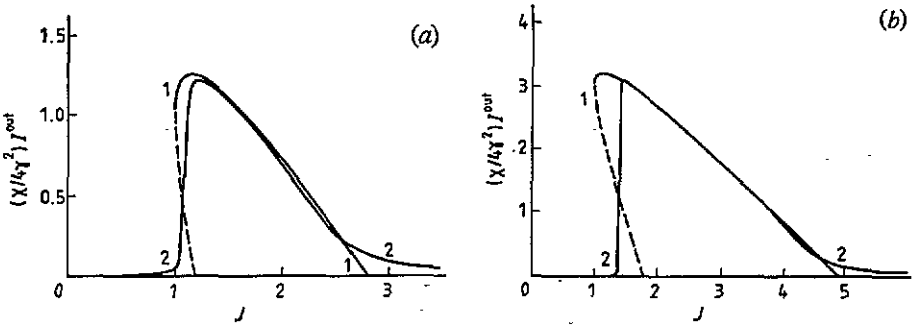

Figure 1. The normalized cavity output intensity of the signal field $(\chi/4\gamma^2)I^{out}$ plotted against the intensity parameter $J = (\chi/\gamma)I$ of the pump fields for $d = 3$ (*a*) and $d = 5$ (*b*). Curves (1) represent the semiclassical result (the broken parts of the curves are related to the unstable solution), while curves (2) represent the exact quantum-mechanical result and they are plotted for $\delta = 0.025$.

Here the first term is the free part of the Hamiltonian, and the last two terms describe the coupling of the signal mode with the reservoir, where Γ and Γ^+ are reservoir operators giving rise to the cavity damping constant γ.

This system in the linear approximation for quantum fluctuations was studied in [13] on the basis of Langevin equations of motion for stochastic field amplitudes. In that paper the stability analysis was carried out for the steady-state semiclassical solutions and the system was shown to produce squeezed light in the above-threshold regime of generation. Note that in the undepleted pump approximation, stable above-threshold generation of the signal field becomes possible only due to inclusion of the self-phase modulation in the model. We list below some of the semiclassical results needed for the following considerations and comparisons.

The semiclassical result for stable steady-state intensity (in units of photon number per unit time) of the signal field at the output of a single-ended cavity in the above-threshold generation regime is [13]

$$I^{out} = 2\gamma\langle a^+a\rangle = \frac{4\gamma^2}{\chi}\left[\frac{\Delta}{\gamma} - \frac{\chi}{\gamma}(I_1 + I_2) + \sqrt{(\chi/\gamma)^2 I_1 I_2 - 1}\right]. \qquad (4)$$

Here $I_{1,2} = |E_{1,2}|^2$ are the intensities of the pump fields (in photon number units), and the well known input–output formalism [14] has been used.

In the case of equal pump intensities ($I_1 = I_2 \equiv I$) the stability domains for above-threshold generation are determined by the following relations

$$I_A < I < I_B \qquad \text{for } \sqrt{3} < \Delta/\gamma < 2 \qquad (5a)$$

and

$$\gamma/\chi < I < I_B \qquad \text{for } \Delta/\gamma > 2. \qquad (5b)$$

Here

$$I_{A,B} = \frac{\gamma}{3\chi}\left(\frac{2\Delta}{\gamma} \mp \sqrt{(\Delta/\gamma)^2 - 3}\right) \qquad (6)$$

where I_A is the threshold value of I. In the region below the generation threshold ($I < I_A$), as well as above I_B, the stability condition is fulfilled only for a zero-amplitude steady-state solution, i.e. the signal field excitation exists at the spontaneous noise level, hence the semiclassical value of the output intensity is $I^{out} = 0$.

The signal field output intensity I^{out} is plotted against the pump intensity in figure 1. The displayed output intensity is essentially bistable in the region of pump intensity from γ/χ to I_A and for detuning $\Delta > 2\gamma$. With increasing detuning parameter Δ/γ the value of I_A, and hence the bistability region, increases. Note that vanishing of the signal field intensity above I_B (in the case $\Delta/\gamma > \sqrt{3}$) is caused by the cross-phase modulation effect. The cross-phase modulation is reflected in the fact that in the equations of motion the usual cavity detuning Δ is replaced by an effective cavity detuning $\Delta_{ef} = \Delta - \chi(|E_1|^2 + |E_2|^2)$ (see equations (11)). This implies that an increase in the pump field intensities $I_{1,2} = |E_{1,2}|^2$ leads to a decrease in Δ_{ef}. As a consequence, the system is carried to the off-resonance operation regime (with respect to the cavity resonance ω_c) and the signal-mode generation vanishes.

It should be pointed out that the linearization procedure about semiclassical steady states is valid if the quantum fluctuations are small. It is clear from the results of [13] that the condition of negligible fluctuations along with the requirement for large mean photon numbers of the signal mode are satisfied if $\chi/\gamma \ll 1$, i.e. for relatively small non-linearities.

Note that almost identical non-linear systems which can be described by an interaction Hamiltonian similar to (2) were studied in [15, 16] in relation to tunnelling and classical amplitude squeezing. The similarity of our model to the model presented in [15] consists in consideration of the parametric driving (with the resulting coupling constant $k \sim \chi^{(2)}E$, where $\chi^{(2)}$ is the second-order susceptibility and E is the amplitude of driving field, or $k \sim \chi^{(3)}E_1E_2$ in our case) combined with Kerr interaction in the form of self-phase modulation. In addition, our model includes the cross-phase modulation effect. However, the main difference of our model from the mentioned models is that in [15, 16] the effects of dissipation for the signal mode and the quantum fluctuations are not included in consideration. It is obvious that if we neglect the cavity damping, the corresponding steady-state solution for the amplitude (intensity) and the phase of the signal mode will not be stable with respect to fluctuations. In addition, the inclusion of the cavity detuning and of the cross-phase modulation affects the specific form of the steady-state solution and leads, in particular, to the appearance of bistability and to the vanishing of the signal mode intensity well above threshold, respectively.

3. Quasiprobability distribution and field moments

We follow the standard procedures (see, for example, [17, 18]) to eliminate the reservoir operators and to obtain a master equation for the density operator ρ of the signal mode. This is readily achieved in an interaction picture relative to the ω_0 frequency, so that the free Hamiltonian, which determines the interaction picture operator evolution, is

$$H_0 = \hbar\omega_0 a^+ a. \tag{7}$$

Thus the interaction picture operators have a time evolution determined by the following transformation to the rotating frame

$$\begin{aligned} a(t) &= a\exp(-\mathrm{i}\omega_c t) \to a\exp(-\mathrm{i}\omega_0 t) \\ a^+(t) &= a^+\exp(\mathrm{i}\omega_c t) \to a^+\exp(\mathrm{i}\omega_0 t). \end{aligned} \tag{8}$$

The resulting master equation in the Markovian approximation and in the interaction picture is

$$\begin{aligned} \frac{\partial\rho}{\partial t} &= \mathrm{i}\Delta[a^+a, \rho] - \tfrac{1}{2}\mathrm{i}\chi(E_1^*E_2^*[a^2, \rho] + E_1E_2[a^{+2}, \rho]) - \tfrac{1}{4}\mathrm{i}\chi[a^{+2}a^2, \rho] \\ &\quad -\mathrm{i}\chi(|E_1|^2 + |E_2|^2)[a^+a, \rho] + \gamma(2a\rho a^+ - \rho a^+a - a^+a\rho). \end{aligned} \tag{9}$$

Here we have assumed that the reservoir temperature satisfies the condition $kT \ll \hbar\omega_0$, hence we neglect thermal fluctuations.

Equation (9) is then easily transformed into a Fokker–Planck equation for the quasiprobability distribution function in the complex [18, 19] P-representation:

$$\frac{\partial}{\partial t}P(\alpha,\alpha^+) = \left[\frac{\partial}{\partial\alpha_\mu}A_\mu + \frac{1}{2}\frac{\partial^2}{\partial\alpha_\mu\partial\alpha_\nu}D_{\mu\nu}\right]P(\alpha,\alpha^+) \tag{10}$$

where $(\alpha,\alpha^+) \equiv (\alpha_1,\alpha_2)$, and, as usual, a summation over the repeating subscripts $\mu, \nu = 1, 2$ is assumed. The drift terms in (10) are

$$A_1 = \gamma\alpha - \mathrm{i}[\Delta - \chi(|E_1|^2 + |E_2|^2)]\alpha + \tfrac{1}{2}\mathrm{i}\chi\alpha^+\alpha^2 + \mathrm{i}\chi E_1E_2\alpha^+ \tag{11a}$$

$$A_2 = \gamma\alpha^+ + \mathrm{i}[\Delta - \chi(|E_1|^2 + |E_2|^2)]\alpha^+ - \tfrac{1}{2}\mathrm{i}\chi\alpha\alpha^{+2} - \mathrm{i}\chi E_1^*E_2^*\alpha \tag{11b}$$

and the diffusion matrix is

$$D = \begin{pmatrix} -\mathrm{i}\chi(E_1E_2 + \frac{1}{2}\alpha^2) & 0 \\ 0 & \mathrm{i}\chi(E_1^*E_2^* + \frac{1}{2}\alpha^{+2}) \end{pmatrix}. \tag{12}$$

We note a multiplicative character of the noise terms, i.e. the amplitude dependence of the diffusion coefficients. At the same time we see that these noise terms grow with increasing non-linearity parameter χ.

The variables α and α^+ in equation (10) are independent complex stochastic c-numbers which correspond to the slowly varying operators a and a^+ such that normally ordered operator moments are obtained via

$$\langle a^{+m}a^n\rangle = \int_C\int_{C'} \mathrm{d}\alpha\,\mathrm{d}\alpha^+\alpha^n\alpha^{+m}P(\alpha,\alpha^+) \tag{13}$$

where C and C' are appropriate integration paths for α and α^+ in the individual (α,α^+) complex planes.

The steady-state solution of equation (10) can be found using the method of potential equations [7, 20]. This yields

$$P_{ss}(\alpha,\alpha^+) = N(\alpha^2 + 2E_1E_2)^\lambda(\alpha^{+2} + 2E_1^*E_2^*)^{\lambda^*}\exp(2\alpha\alpha^+) \tag{14}$$

where

$$\lambda = -1 - 2\Delta/\chi + 2(|E_1|^2 + |E_2|^2) - 2\mathrm{i}\gamma/\chi \tag{15}$$

and N is the normalization constant.

Using the solution (14) and noting that the integrals in (13) are identical to those in the definition of the betafunction [21]

$$\int_C t^a(t^2-1)^b\mathrm{d}t = \begin{cases} 0 & \text{for odd } a \\ 2\mathrm{i}\sin(\pi b)B\left(\dfrac{a+1}{2}, b+1\right) & \text{for even } a \end{cases} \tag{16}$$

where C is an eight-shaped contour encircling the points $t = \pm 1$, we obtain for all the normally-ordered moments of the signal field operators:

$$\langle a^{+m}a^n\rangle = M_{mn}/M_{00} \tag{17}$$

$$M_{mn} = (-2E_1E_2)^{n/2}(-2E_1^*E_2^*)^{m/2}\sum_{k=0}^{\infty}\frac{|4E_1E_2|^k}{k!}[1+(-1)^{k+n}][1+(-1)^{k+m}]$$
$$\times B\left(\lambda+1, \frac{k+n+1}{2}\right)B\left(\lambda^*+1, \frac{k+m+1}{2}\right). \tag{18}$$

4. Exact steady-state intensity

Using the general results (17) and (18), and transforming the betafunctions to gammafunctions ($B(x, y) = \Gamma(x)\Gamma(y)/\Gamma(x+y)$), we write down the steady-state intensity of the cavity output signal field:

$$I^{\mathrm{out}} = 2\gamma\langle a^+ a\rangle = 2\gamma M'_{11}/M'_{00} \tag{19}$$

$$M'_{00} = \frac{1}{1+(J_1+J_2-d-\delta)^2} + \sum_{n=1}^{\infty}\frac{(2n-1)!!}{(2n)!!}\frac{(J_1J_2)^n}{\prod_{k=1}^{n+1}[1+(J_1+J_2-d-3\delta+2k\delta)^2]} \tag{20}$$

$$M'_{11} = \frac{J_1J_2}{2}\sum_{n=0}^{\infty}\frac{(2n+1)!!}{(2n)!!}\frac{(J_1J_2)^n}{\prod_{k=1}^{n+2}[1+(J_1+J_2-d-3\delta+2k\delta)^2]}. \tag{21}$$

Here we have used the moments M'_{mn}, which differ from the M_{mn} by a constant factor

$$M_{mn} = \frac{16\pi\delta^2|\lambda|^2\Gamma(\lambda)\Gamma(\lambda^*)}{\Gamma(\lambda+\frac{1}{2})\Gamma(\lambda^*+\frac{1}{2})}M'_{mn} \tag{22}$$

and we have defined

$$J_{1,2} = \frac{\chi}{\gamma}I_{1,2} \qquad d = \frac{\Delta}{\gamma} \qquad \delta = \frac{\chi}{4\gamma}. \tag{23}$$

We present here the results of numerical calculations for the case of equal pump intensities $I_1 = I_2 = I$. The normalized output intensity of the signal field is plotted in figure 1 as a function of the pump intensity parameter $J = (\chi/\gamma)I$ for different values of the detuning parameter $d = \Delta/\gamma$. An increase in d leads to extension of the generation region, where the output intensity differs essentially from zero, as well as to an increase in the maximal value of the signal intensity and to an increase in the threshold values of the pump intensity. Note also that with increasing the relative non-linearity parameter δ, the intensity of the signal mode in the threshold region increases more smoothly, i.e. the transition region becomes broader.

The absence of hysteresis-cycle behaviour of the exact quantum mechanical mean intensity, which takes into account quantum fluctuations and has a strictly statistical sense, is a result of well known studies on optical bistability (see, for example, [22] and references therein) and, of course, is a fact of general character. Correspondingly, in a quantum statistical treatment it is more adequate to speak of metastable states connected with a probability (quasiprobability) distribution function (or with a generalized potential) instead of stable semiclassical steady states. In this case in order to describe the bistability phenomenon in more detail one must deal with a transient behaviour and with the approach of a particular non-linear system to a steady state from a statistical viewpoint. As a result, one can speak of characteristic transition times (or quantum tunnelling times) between the metastable states and one can estimate these times in order to ensure the reliability of a bistable device on the usual laboratory time scales. This problem, however, needs special consideration for our non-linear system and its solution is beyond the framework of the present paper.

5. Second-order correlation function

The normalized second-order correlation function is defined as

$$g^{(2)}(0) = \frac{\langle a^+a^+aa\rangle}{\langle a^+a\rangle^2} = \frac{M'_{22}M'_{00}}{(M'_{11})^2} \tag{24}$$

where M'_{00} and M'_{11} are given by equations (20) and (21) and the second-order moment M'_{22} takes the form

$$M'_{22} = \frac{J_1 J_2}{4} \sum_{n=0}^{\infty} \frac{(2n+1)!!}{(2n)!!} \frac{(2n+1)(J_1 J_2)^n}{\prod_{k=1}^{n+2}[1 + (J_1 + J_2 - d - 3\delta + 2k\delta)^2]}. \tag{25}$$

The result of numerical calculation of the correlation function (24) is plotted against the pump intensity parameter $J = (\chi/\gamma)I$ (for $I_1 = I_2 \equiv I$) in figure 2. It is readily seen that below the critical transition region the correlation function shows superbunching behaviour. This feature is caused by pair creation of the signal mode photons in the FWM process. With increasing parameter J the correlation function decreases, however in the critical threshold region $g^{(2)}(0)$ has a sharp peak indicating a critical increase in the quantum fluctuations: $g^{(2)}(0) = 1 + \langle : (\Delta n)^2 : \rangle / \langle n \rangle^2 \gg 1$, where $\langle : (\Delta n)^2 : \rangle$ is the normally-ordered dispersion of photon number fluctuations, $\langle n \rangle = \langle a^+ a \rangle$ is the mean photon number. This peak is well localized for small values of the parameter $\delta = \chi/4\gamma$, however it becomes both smaller and broader as the relative non-linearity χ/γ increases and it disappears for $\chi/\gamma \sim 1$. Hence the characteristic threshold behaviour, defined by a drastic increase in the photon number fluctuations in the transition region, disappears in the case of strong non-linearities. As can be seen from equation (12) for diffusion coefficients, such a behaviour corresponds to a dramatic increase in the role of quantum noise.

In the above-threshold region, where the signal field intensity grows substantially, the self-phase modulation effect becomes essential and the correlation falls, becoming smaller than unity, although remaining close to it. This implies a minor non-classical effect of photon antibunching. Note that such behaviour is in qualitative agreement with the result obtained in [7] for the process of pure self-phase modulation in a coherently driven cavity. In terms of the Fano parameter

$$F = \langle (\Delta n)^2 \rangle / \langle n \rangle = 1 + \langle n \rangle (g^{(2)} - 1) \tag{26}$$

this leads to sub-Poissonian photon statistics when a reduction of photon number fluctuations below the coherent level ($F < 1$) occurs. For example, in the case of $d = 5$ and $\delta = 0.01$ the minimal value of the Fano factor is $F \simeq 0.82$, and this value decreases slightly with decreasing δ ($F \simeq 0.67$ in the case $\delta = 0.001$). In contrast, as δ increases the Fano factor becomes $F \sim 1$ and the sub-Poissonian statistics disappear.

With further increase in the pump intensity parameter J the antibunching effect disappears and it is replaced by photon bunching ($g^{(2)}(0) > 1$). This behaviour is related to the fact, that, far above threshold ($I > I_B$), the system returns to the generation regime at the spontaneous noise level.

6. Summary

An exact quantum treatment is presented here for the parametric four-wave mixing interaction of two pump fields of different frequencies with an intracavity signal mode of degenerate frequency ($\omega_1 + \omega_2 \to 2\omega_c$). The model considered incorporates self- and cross-phase modulation effects of the signal mode, which would normally arise in a $\chi^{(3)}$-medium.

An exact steady-state solution of the Fokker–Planck equation for the quasiprobability P-function was obtained. This solution allowed us to write down an analytic expression for arbitrary moments of the signal field operators. The behaviour of the exact steady-state intensity and quantum-statistical properties of the signal field were studied in detail.

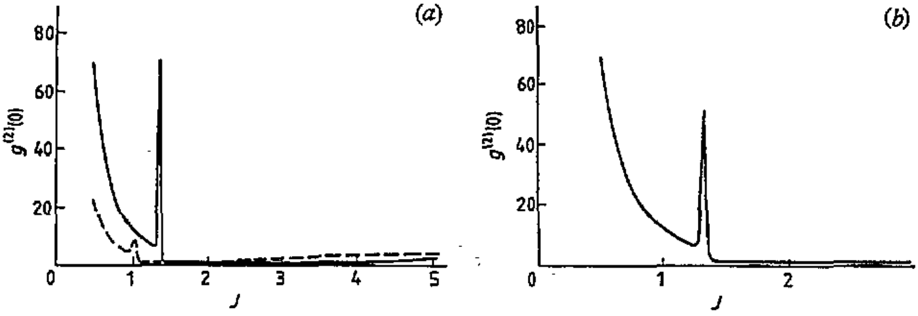

Figure 2. The second-order correlation function $g^{(2)}(0)$ plotted against $J = (\chi/\gamma)I$: (*a*) $\delta = 0.025$, the full curve corresponds to $d = 5$, while the broken curve corresponds to $d = 3$; (*b*) $\delta = 0.04$ and $d = 5$.

The dependence of the signal field intensity on the χ/γ dimensionless parameter of non-linearity shows a broadening of the transition region in the case of large non-linearities ($\chi/\gamma \sim 1$), in contrast to the $\chi/\gamma \ll 1$ case, when the intensity grows drastically. At the same time, the behaviour of the second-order correlation function and of the photon number fluctuations in this transition region shows a well localized sharp peak in the case of small non-linearities ($\chi/\gamma \ll 1$). Localization of this peak can be identified with a threshold in the quantum statistical treatment. However, this characteristic peak becomes smaller and broader as χ/γ increases, and it disappears for $\chi/\gamma \sim 1$.

Above the transition region, where the signal field intensity grows substantially, the photon statistics displays antibunching and sub-Poissonian behaviour. With a further increase in the pump intensity the system returns to the regime of generation at the spontaneous noise level, and the second-order correlation function shows photon bunching.

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