## Sub-Poissonian Photon Statistics in a Three-Wave Interaction Starting in the Out-of-Phase Regime

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We present for the first time numerical and analytical calculations for the nonlinear interaction of three quantized waves all sizably excited from the beginning and having different phase relations. With a Kerr-state ansatz for the signal we get strongly sub-Poissonian photon statistics and conclude on similar effects by initially entangled states.

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Nonlinear interaction between light waves has been studied since the laser came into existence in 1960. Shortly after this a comprehensive paper appeared [1] that describes three-wave and four-wave interactions in nonlinear media and gives the exact solutions for the classical coupled-mode equations. This work is now at the heart of nonlinear optics and the basis of many quantum optical investigations. Let us confine ourselves to the three waves, i.e.,  $\chi^{(2)}$  media, and consider the general case that all waves are excited at the beginning of the interaction. We limit ourselves to exact resonance  $\omega_3 = \omega_1 + \omega_2$ , where  $\omega_3$  is the frequency of the pump and  $\omega_1$  and  $\omega_2$  are the frequencies of the signal and idler, respectively. In addition, we assume phase matching of the wave vectors, which provides the justification for retaining only these three waves or modes.

An important approximation that allows an analytic solution also in the quantum case is the so-called parametric one, where the strong pump wave is treated as a  $c$  number and any depletion is neglected, while the relatively weak signal and idler can change considerably [2]. This approximation is easily applied but its justification is hard to show [3]. Most quantum optical investigations with  $\chi^{(2)}$ media are carried out in this approximation [4].

However, an arbitrary preparation of all three waves is now experimentally possible with different phase relations between them. Classically the solutions are well known, but quantum mechanically we have to resort to numerical methods [5]. In their first calculations Walls and Barakat started with Fock states, i.e., without setting any phase relation. By now these initial states have been replaced by coherent states, but at least one mode has still been in the vacuum. This vacuum mode then becomes excited and its phase adjusts automatically to the right phase difference. Thus also with two excited coherent states the manifold of phase relations cannot be explored.

Here we present for the first time numerical and analytical calculations for the quantum case, where all three modes are considerably excited from the beginning, and choose a particular phase relation between them. A coherent state with a mean photon number exceeding 5 has already a sufficiently sharp mean phase [6]. Hence starting with such states in all three modes allows one, therefore, to choose an arbitrary initial phase relation between them for nondegenerate three-wave interaction. This is compared with the classical solutions and applied to a specially prepared signal, which under sum-frequency generation then shows strong sub-Poissonian photon statistics.

We start with the Hamiltonian for the nondegenerate three-wave interaction in the interaction picture

$$
\hat{H}_{\text{int}} = \hbar \kappa (\hat{a}\hat{b}\hat{c}^{\dagger} + \hat{a}^{\dagger}\hat{b}^{\dagger}\hat{c}), \qquad (1)
$$

where  $\hat{c}$  ( $\hat{c}^{\dagger}$ ) is the annihilation (creation) operator of the pump mode and  $\hat{a}$  and  $\hat{b}$  denote the corresponding operators for the signal and idler, respectively. The coupling constant  $\kappa$  contains the nonlinear susceptibility  $\chi^{(2)}$ . This Hamiltonian (1) describes phase-dependent and phase-insensitive amplification including saturation, sum-frequency generation, and frequency conversion. In addition, (1) is also the interaction for special quantum effects as two-mode squeezing and correlated beams for various interference experiments. Even the Jaynes-Cummings model is a special case of (1). So any new effect discovered in (1) can extend our understanding of this fundamental Hamiltonian.

Because the system (1) can only be numerically solved, we limit our analytical treatment to a short-time expansion. For the signal mode operator we find from the Heisenberg equations of motion

$$
\hat{a}(t) = \hat{a} - i\kappa t \hat{b}^\dagger \hat{c} + \frac{(\kappa t)^2}{2!} (\hat{c}^\dagger \hat{a} \hat{c} - \hat{b}^\dagger \hat{a} \hat{b}) + O((\kappa t)^3),
$$
\n(2)

where all operators without a time argument are taken at  $t = 0$ . Equivalent expressions can be written for  $\hat{b}$  and  $\hat{c}$ . The signal photon number is then given by

$$
\hat{a}^{\dagger}(t)\hat{a}(t) = \hat{a}^{\dagger}\hat{a} + i\kappa t(\hat{c}^{\dagger}\hat{b}\hat{a} - \hat{a}^{\dagger}\hat{b}^{\dagger}\hat{c})
$$

$$
+ (\kappa t)^{2}[\hat{c}^{\dagger}\hat{c}(\hat{a}^{\dagger}\hat{a} + \hat{b}\hat{b}^{\dagger}) - \hat{a}^{\dagger}\hat{a}\hat{b}^{\dagger}\hat{b}]
$$

$$
+ O((\kappa t)^{3}). \tag{3}
$$

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Assume now initially coherent states in all three modes

$$
|\alpha\rangle_a|\beta\rangle_b|\gamma\rangle_c \quad (\alpha = |\alpha|e^{i\varphi_a}, \beta = |\beta|e^{i\varphi_b}, \gamma = |\gamma|e^{i\varphi_c}), \tag{4}
$$

then the expectation value of (3) results in

$$
\langle \hat{a}^{\dagger}(t)\hat{a}(t)\rangle = |\alpha|^2 + 2\kappa t |\alpha||\beta||\gamma| \sin(\Delta\varphi) + \kappa^2 t^2 \times [|\gamma|^2(|\alpha|^2 + |\beta|^2 + 1) - |\alpha|^2|\beta|^2] + O(\kappa^3 t^3). \tag{5}
$$

We call the linear terms in  $\kappa t$  the coherent ones because they depend on the phase difference. If  $\Delta \varphi =$  $\varphi_c - \varphi_b - \varphi_a = -\pi/2$ , the signal starts getting attenuated, while for  $\Delta \varphi = \pi/2$  the signal is first amplified. Both values are distinguished insofar as the phase difference stays with them both quantum mechanically and classically as can be seen in the following. In addition, we consider the phase difference  $\Delta \varphi = 0$ , where all phasedependent terms in (5) disappear and the direction of the process is given simply by the intensity relations (second order term). We call this initial condition the out-of-phase regime. Note further that the fluctuations enter (3) in the second-order term by the commutation relation of the idler operators. We need also the squared number operator

$$
[\hat{a}^{\dagger}(t)\hat{a}(t)]^2 = \hat{a}^{\dagger 2}\hat{a}^2 + \hat{a}^{\dagger}\hat{a} + i2\kappa t(\hat{a}^{\dagger}\hat{a}^2\hat{c}^{\dagger}\hat{b} - \hat{a}^{\dagger 2}\hat{a}\hat{b}^{\dagger}\hat{c})
$$

$$
+ i\kappa t(\hat{a}\hat{c}^{\dagger}\hat{b} - \hat{a}^{\dagger}\hat{b}^{\dagger}\hat{c}) + O(\kappa^2 t^2), \quad (6)
$$

where we can confine ourselves to the first order of time effects.

For three coherent states the disappearance of the first order terms in (5) would result in the same effect in the expectation value of (6). However, for more general states, we can put these terms to zero in the photon number and nevertheless retain nonzero terms for the expectation value of (6). This could give a strong tendency to sub-Poissonian statistics. Note two possibilities for this in (6). First, there are higher moments of  $\hat{a}$ ,  $\hat{a}^{\dagger}$  and second, due to some entanglement, any factorization of the expectation values of the different modes may become impossible. We will discuss both properties but focus on the first. Eventually we should add that this short-time expansion approach is by no means new [7]. However, earlier investigations gave only second order of time contributions to the fluctuations of signal or idler becoming so super-Poissonian. Our approach is to show that there can be first order of time terms that drive the signal to strong sub-Poissonian statistics.

Because the case  $\Delta \varphi = 0$  (no coherent interaction at the beginning) is normally avoided even in the classical coupled-mode equations [1], we should briefly discuss the classical interaction. Introducing a scaled time by  $\zeta = \kappa t$ we get for the classical amplitudes the equations

$$
\frac{d u_1}{d \zeta} = -u_2 u_3 \sin \theta ,
$$
  
\n
$$
\frac{d u_2}{d \zeta} = -u_1 u_3 \sin \theta ,
$$
  
\n
$$
\frac{d u_3}{d \zeta} = u_2 u_1 \sin \theta ,
$$
\n(7)

where  $u_1, u_2, u_3$  are the classical (slowly varying) amplitudes of the signal, idler, and pump, respectively, and  $\theta(\zeta) = \phi_3(\zeta) - \phi_2(\zeta) - \phi_1(\zeta)$ . The equation of motion for  $\theta(\zeta)$  is

$$
\frac{d\theta}{d\zeta} = \frac{\cos\theta}{\sin\theta} \frac{d}{d\zeta} \ln(u_1 u_2 u_3).
$$
 (8)

We neglect here any mismatch and mention three Manley-Rowe relations  $m_1 = u_2^2 + u_3^2$ ,  $m_2 = u_1^2 + u_3^2$ ,  $m_3$  $u_1^2 - u_2^2$  that have their corresponding conserved quantities in the quantum system described by (1). Thus we can interpret  $u_1^2, u_2^2, u_3^2$  as photon numbers in the corresponding modes if we introduce a suitable rescaling of the  $u_i$  and  $\zeta$  [8] that does not change the form of the equations. Hence (7) and (8) are the classical equivalents of the Heisenberg equations of motion derived from (1).

The advantage of (8) is that it can immediately be integrated to give

$$
u_1(\zeta)u_2(\zeta)u_3(\zeta)\cos[\theta(\zeta)] = \Gamma
$$
  
=  $u_1(0)u_2(0)u_3(0)\cos[\theta(0)].$   
(9)

Note, however, that there are single equations for each phase which are not solved by this approach [9]. As an example, the equation for  $\phi_1(\zeta)$  reads  $d \phi_1(\zeta)/d \zeta =$  $(u_2u_3/u_1)\cos\theta$  and proves the change of  $\phi_1$  for  $\cos\theta \neq$ 0. With the help of (9) and the Manley-Rowe relations the system (7) can then be integrated, which results in the well-known Jacobian elliptic functions for the  $u_i^2(\zeta)$  $(i = 1, 2, 3)$ .

The solutions of (7) depend decisively on  $\Gamma$  given in (9), and a cubic equation containing  $\Gamma$  has to be solved. For  $\theta(0) = 0$  (equivalent to  $\Delta \varphi = 0$ ) this is done via a quadratic equation. The mutual relations of the three roots determine whether the signal first increases or not for  $\theta(0) = 0$ . Note that then the phase difference changes in the direction that favors the energy exchange. Simultaneously the single phases change much faster [9].

The quantum mechanical equivalent of (9) is the expectation value of the interaction Hamiltonian (1). Hence the subtle classical results have their quantum mechanical analogs in the solution of (1). A detailed discussion of it [9] shows also why the phase difference stays at its value  $\Delta \varphi = \pm \pi/2$  both in the classical as well as in the quantum description and each single phase keeps its constant value.

Here we illustrate these facts by our numerical results and then show the nonclassical features. Figure 1(a)



FIG. 1. (a) Change of the signal mean photon number for the initial state (4) with  $\Delta \phi = 0$  and  $\Delta \phi = -\pi/2$ . The full lines. represent the quantum calculation of (1), while the dashed lines show the classical solution of (7) and (8). (b)  $Q$  parameter (as defined in the text) of the signal (joined) and idler for the initial state (4).

demonstrates the behavior of the signal photon number for the initial state (4) with  $|\alpha| = 6$ ,  $|\beta| = 4$ ,  $|\gamma| =$ 3. Note that the period of the energy exchange is almost the same as for both  $\Delta \varphi = 0$  and  $-\pi/2$  even though these amplitude parameters prevent a sizable energy transfer for  $\Delta \varphi = 0$ . Both processes start with sumfrequency generation, but the  $Q$  parameter of the signal,  $Q = \left[ \langle (\hat{a}^\dagger \hat{a})^2 \rangle - \langle \hat{a}^\dagger \hat{a} \rangle^2 \right] / \langle \hat{a}^\dagger \hat{a} \rangle - 1$ , plotted in Fig. 1(b) goes negative only after the second start of sum-frequency generation, indicating strongly sub-Poissonian behavior of the signal in the case  $\Delta \varphi = -\pi/2$ . This striking effect is the central point of our investigation. Analogous effects were observed with the initial conditions  $|\alpha\rangle_a|0\rangle_b|\gamma\rangle_c$ ,  $|\alpha\rangle_a|\beta\rangle_b|0\rangle_c$ , where the initial coherent amplitudes could be as high as  $\alpha = \gamma = 8$  starting with difference-frequency generation [10] and  $\alpha = 9, \beta = 5$  generating first the sum frequency [11]. The phases in the newly generated modes adjust in such a way that corresponds to  $\Delta \varphi = \pm \pi/2$ , respectively.

Achieving a remarkable nonclassical effect after one quasiperiod in three-wave interaction is physically very interesting, but experimentally still unrealistic. Therefore we tried to understand the mechanism of this effect and to find ways to realize it within the first quasiperiod. One method to get this without any entanglement at the beginning is a Kerr-state ansatz for the signal mode and coherent states for the idler and pump. This uses the properties of a Kerr state [12] that

$$
K \langle \alpha | \hat{a}^{\dagger 2} \hat{a} | \alpha \rangle_K = \alpha^* |\alpha|^2 e^{-|\alpha|^2 (1 - \cos \epsilon) + i |\alpha|^2 \sin \epsilon + i \epsilon}, \quad (10)
$$

$$
\langle \langle \alpha | \hat{a}^{\dagger} | \alpha \rangle_K = \alpha^* e^{-|\alpha|^2 (1 - \cos \epsilon) + i |\alpha|^2 \sin \epsilon}, \qquad (11)
$$

where  $|\alpha\rangle_K$  is a Kerr state developed from a coherent state and  $\kappa \langle \alpha | \hat{a}^{\dagger} | \alpha \rangle_K = \langle \alpha | \hat{a}^{\dagger} e^{i \epsilon \hat{a}^{\dagger} \hat{a}} | \alpha \rangle$ . In these equations is a parameter that contains  $\chi^{(3)}$  and the interaction length. If we compare (10) and (11) the small phase shift  $\epsilon$  introduced by the commutation relations becomes important and leads for

$$
\varphi_a + \varphi_b - \varphi_c - |\alpha|^2 \sin \epsilon = 0 \qquad (12)
$$

to a nonzero first-order term in the expectation value of (6), while the photon number has no such contribution. The variance of the photon number is then

$$
\langle [\hat{a}^{\dagger}(t)\hat{a}(t)]^2 \rangle - \langle \hat{a}^{\dagger}(t)\hat{a}(t) \rangle^2 = |\alpha|^2
$$
  
+ 4\kappa t |\alpha|^3 |\beta| |\gamma| sin \epsilon  
+ O(\kappa^2 t^2). (13)

This equation shows for  $\epsilon < 0$  a strong tendency to sub-Poissonian statistics.

The disappearance of the first order terms in (5) with the Kerr state in the signal at the phase difference (12) is equivalent to a vanishing field strength in the Kerr state, because the coherent pump and idler define a reference phase for the signal. This is exactly the condition for getting extreme sub-Poissonian statistics by interference of a coherent beam and a Kerr-state field [13].

In Fig. 2(a) we plot the  $Q$  parameter of the signal for the interaction (1) and the initial state  $||\alpha| = 6$ <sub>K</sub> $||\beta|$  =  $4\rangle_b ||\gamma| = 3\rangle_c$  imposing the phase relation (12). Already a very short interaction time is sufficient to result in strong sub-Poissonian photon statistics. On the other hand, the extremes obtained by interference [12,13] cannot be reached because the phases move classically in this configuration out of the position  $\Delta \varphi = 0$ . This is illustrated in Fig. 2(b). We should mention that we get a similar result if we start with  $||\alpha| = 4$ <sub>K</sub> $||\beta| = 3$ <sub>b</sub> $||\gamma| = 6$ <sub>c</sub>, but now with initial amplification of the signal for  $\Delta \varphi = 0$ .

Finally we return to the result in Figs. 1(a) and 1(b). There is, of course, no Kerr effect at the restart of sumfrequency generation. But our Kerr-state calculations



FIG. 2. (a) The  $Q$  parameter of the signal as a function of time for the Kerr-state ansatz in the signal and coherent states in the idler and pump. The amplitudes are  $|\alpha| = 6$ ,  $|\beta| =$ 4,  $|\gamma| = 3$  and the phases fulfill Eq. (12) with  $\epsilon = -0.1$ . (b) Outer Q-function contour lines (height 0.01) of the signal at scaled times  $\kappa t = 0.0, 0.2, 0.4, 0.6$  for the initial state (4) with  $\Delta \varphi = 0$ . The signal starts with a circular contour line. The corresponding classical motion of the signal (up to the scaled time 0.6) is plotted (in polar coordinates) in the insertion and starts on the positive real axis.

teach us that there must be a first-order of time reduction effect of the photon number variance. However, this is not conceivable in a disentangled state, because at this moment we have  $\langle \hat{c} \rangle = \langle \hat{c}^{\dagger} \rangle = 0$ ; i.e., the coherent pump amplitude crosses zero. That the first-order terms in the photon number vanish and only the first-order terms in the expectation value of (6) survive follows also analytically because the first derivative of the signal and idler photon number has to vanish here. Hence the entanglement between signal and pump is responsible for this strong tendency to sub-Poissonian photon statistics in Fig. 1(b), which is discussed in more detail in [11]. This entanglement is generated in the first quasiperiod in Fig. 1(a), but it might be possible to generate it in a different way in order to use it as signal-pump initial state. Note that the signal-pump entanglement is the result of a saturated amplification process and therefore a fundamental property of this amplifier model. In this way it offers the possibility to learn more about saturated amplifiers. Moreover, we found [10] that during the following sum-frequency generation the signal entanglement is strongly reduced so that the signal ends up in almost a pure state with strong sub-Poissonian behavior. This corroborates our interpretation with the signal-pump entanglement and shows that it can be used up.

In addition, we should concede that the nonclassical character of the Kerr state  $(10)$  and  $(11)$  promotes the occurrence of the sub-Poissonian character in Fig. 2(a). However, at the start of the interaction (1) there is only Poissonian photon statistics and no amplitude squeezing because the expectation value of the Kerr field strength vanishes at the phase position (12). A normal simple absorption process would never result in such nonclassical properties but on the contrary destroy them. In such a way the depletion in signal and idler by sum-frequency generation is similar to an interference process. A last remark concerns the phase relation (12). The depletion process would be phase stable for  $\Delta \phi = -\pi/2$  but then the Kerr state could only deliver an effect proportional to  $\epsilon^2$  in (13), while for (12) it is of the order  $\epsilon$ .

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