Optica Applicata, Vol. IX, No. 4, 1979

On the efficiency of nonlinear optical processes with respect to photon statistics of generating radiations*

PAVEL CHMELA
Palacký University, Laboratory of Optics, Olomouc, Czechoslovakia.

The effect of photon statistics on the course of nonlinear optical three-mode processes is discusse in this paper. A method that assumes conserved statistics of generating radiations in the course of a nonlinear process [8] is used and two typical cases, namely the parametric generation from quantum noise and parametric up-conversion, are studied here. The influence of photon statistics of generating radiations upon the course of nonlinear optical processes is explained by the effects of intermodal correlations.

Introduction

An optical non-degenerate three-mode nonlinear process in a non-dissipative medium can be described by means of the trilinear time-dependent Hamiltonian [1–5]:

$$H(t) = \hbar[\omega_1 a_1^+(t)a_1(t)+\omega_2 a_2^+(t)a_2(t)+\omega_3 a_3^+(t)a_3(t)]+ \\ +\hbar g[a_1(t)a_2(t)a_3^+(t)+a_1^+(t)a_2^+(t)a_3(t)], \qquad (1)$$

where ω_1, ω_2, ω_3 are the frequencies of considered modes satisfying the condition:

$$\omega_1+\omega_2 = \omega_3; \qquad (2)$$

$a_i(t)$, $a_i^+(t)$ label the annihilation and creation operators relative to the i-th mode, and g is the real coupling constant.

The phase matching for respective wave vectors is assumed as well, i.e.

$$\boldsymbol{k}_1+\boldsymbol{k}_2 = \boldsymbol{k}_3. \qquad (3)$$

The differential equations for the annihilation operators can be obtained when using the Heisenberg equation of motion [1–5]:

$$i\frac{da_1}{dt} = \omega_1 a_1+ga_2^+a_3, \qquad (4a)$$

$$i\frac{da_2}{dt} = \omega_2 a_2+ga_1^+a_3, \qquad (4b)$$

$$i\frac{da_3}{dt} = \omega_3 a_3+ga_1a_2. \qquad (4c)$$

* This paper has been presented at the Fourth Polish-Czechoslovakian Optical Conference in Rynia (near Warsaw), September, 19–22, 1978.

Solution of the equations (4) represents the well known three-body problem and thus a solution in a closed form does not seem possible. The equations (4) were solved in the short-time approximation by Agrawal [4] and Peřina [5, 7], and similar equations for a degenerate process were solved by Kielich [6] and Peřina [7].

In some special cases it seems to be advantageous to use a method proposed by Crosignani [8] which assumes that the photon statistics of generating radiations is not changed in the course of the nonlinear process. Such an assumption allows to find the solution of (4) in a closed form. It is interesting that for second harmonic generation, for which the method was originally proposed, this method seems to offer very low accuracy.

In this paper we shall consider two cases, namely the parametric generation from quantum noise and the parametric up-conversion.

The basic relations, we shall use here, are the equations of motion for photon number operators $n_i(t) = a_i^+(t)\, a_i(t)$ following from (4):

$$\frac{d^2 n_1}{dt^2} = \frac{d^2 n_2}{dt^2} = -\frac{d^2 n_3}{dt^2} = 2g^2(n_3 n_1 + n_3 n_2 - n_1 n_2 + n_3), \tag{5}$$

and the photon number conservation laws:

$$\frac{dn_1}{dt} - \frac{dn_2}{dt} = 0, \tag{6a}$$

$$\frac{dn_1}{dt} + \frac{dn_3}{dt} = 0, \tag{6b}$$

$$\frac{dn_2}{dt} + \frac{dn_3}{dt} = 0. \tag{6c}$$

In order to solve the averaged equation (5) it is necessary to assume the factorization relations for generating radiations of the type $\langle n_i^2(0)\rangle$, $\langle n_i^2(t)\rangle$, $\langle n_i(t)\, n_i(0)\rangle$ and to introduce the corresponding initial conditions.

We shall consider two types of generating radiations:

a. Coherent generating radiations with Poisson photon number distribution satisfying the factorization relations of the type [9]:

$$\langle n_i^2(0)\rangle = \langle n_i(0)\rangle^2 + \langle n_i(0)\rangle, \tag{7a}$$

$$\langle n_i^2(t)\rangle = \langle n_i(t)\rangle^2 + \langle n_i(t)\rangle, \tag{7b}$$

$$\langle n_i(t)\, n_i(0)\rangle = \langle n_i(t)\rangle \langle n_i(0)\rangle + \langle n_i(t)\rangle. \tag{7c}$$

b. Chaotic (Gaussian) generating radiations with Bose–Einstein statistics that satisfy the factorization relations of the type [9]:

$$\langle n_i^2(0)\rangle = 2\langle n_i(0)\rangle^2 + \langle n_i(0)\rangle, \tag{8a}$$

$$\langle n_i^2(t)\rangle = 2\langle n_i(t)\rangle^2 + \langle n_i(t)\rangle, \tag{8b}$$

$$\langle n_i(t)\, n_i(0)\rangle = [1+\exp(-2\Gamma_i|t|)]\langle n_i(t)\rangle\langle n_i(0)\rangle + \langle n_i(t)\rangle, \tag{8c}$$

where Γ_i denotes the spectral half-width of Lorentzian line.

Parametric generation from quantum noise

Parametric generation from quantum noise is a spontaneous decay of a pumping photon at ω_3 into two subfrequency photons at ω_1 and ω_2.

From the classical theory the zero effect can be derived.

The quantum theory has shown that the parametric generation from quantum noise can start because of quantum fluctuations.

The parametric generation from quantum noise is characterized by the following initial conditions:

$$\langle n_1(0)\rangle = \langle n_2(0)\rangle = 0;\ \langle n_3(0)\rangle = n_{3,0}; \tag{9a}$$

$$\left\langle\left(\frac{dn_1}{dt}\right)_{t=0}\right\rangle = \left\langle\left(\frac{dn_2}{dt}\right)_{t=0}\right\rangle = \left\langle\left(\frac{dn_3}{dt}\right)_{t=0}\right\rangle = 0. \tag{9b}$$

a. For the coherent pumping radiation it has been found that when assuming quite intense pumping radiation, i.e. $n_{3,0} \gg 1$, the following expressions for mean photon numbers in the individual modes hold [10]:

$$\langle n_1(t)\rangle = \langle n_2(t)\rangle = n_{3,0}\frac{\tan h^2(n_{3,0}^{1/2}gt)}{[1+n_{3,0}\sec h^2(n_{3,0}^{1/2}gt)]}, \tag{10a}$$

$$\langle n_3(t)\rangle = \qquad = n_{3,0}\frac{(n_{3,0}+1)\sec h^2(n_{3,0}^{1/2}gt)}{[1+n_{3,0}\sec h^2(n_{3,0}^{1/2}gt)]}. \tag{10b}$$

The average time of the first photon decay was found to be [10]:

$$\langle\tau_{\mathrm{phot}}\rangle_{\mathrm{coh}} \doteq \frac{0.88}{n_{3,0}^{1/2}g}. \tag{11}$$

It follows from (10) that the total conversion of pumping radiation energy at ω_3 into the energy of two subfrequencies ω_1 and ω_2 can take place as t goes to infinity, provided that $n_{3,0} \gg 1$. However, the time of the total energy conversion is very long when compared with other nonlinear processes.

b. For chaotic pumping radiation the parametric generation from quantum noise does start, if the coherence time ($\tau_{c,3}$) is exteremely long and the condition

$$\tau_{c,3} = \frac{1}{2\Gamma_3} > \frac{(2n_{3,0})^{1/2}}{g} \tag{12a}$$

hold [10].

In the opposite case, if

$$\tau_{c,3} = \frac{1}{2\Gamma_3} \lesssim \frac{(2n_{0,3})^{1/2}}{g}, \tag{12b}$$

the process does not start at all [10].

The condition (12a) could be fulfilled for pumping radiations with extremely high coherence degree. However, such radiation are not emitted by usual thermal sources.

Parametric up-conversion

The parametric up-conversion means a nonlinear process of sum-frequency generation at ω_3, the intensity of one subfrequency component at ω_1 (pumping radiation) being essentially greater than the intensity of the other one at ω_2 (input radiation). This process is characterized by the initial conditions:

$$\langle n_1(0)\rangle = n_{1,0};\ \langle n_2(0)\rangle = n_{2,0};\ \langle n_3(0)\rangle = 0; \tag{13a}$$

$$\left\langle\left(\frac{dn_1}{dt}\right)_{t=0}\right\rangle = \left\langle\left(\frac{dn_2}{dt}\right)_{t=0}\right\rangle = \left\langle\left(\frac{dn_3}{dt}\right)_{t=0}\right\rangle = 0, \tag{13b}$$

while the following relations hold:

$$n_{1,0} \gg n_{2,0};\ n_{1,0} \gg \langle n^2(t)\rangle;\ n_{1,0} \gg \langle n_3(t)\rangle. \tag{14}$$

It has been shown in [11] that the efficiency of parametric up-conversion is practically insensible to photon statistics of the input radiation at ω_2. The description, that is used here, corresponds rather to the classical one [11].

a. For the coherent pumping radiation at ω_1 the following approximative expressions for mean photon numbers in the individual modes have been found [11].

$$\langle n_1(t)\rangle = n_{1,0} - n_{2,0}\sin^2(n_{1,0}^{1/2}gt), \tag{15a}$$

$$\langle n_2(t)\rangle = n_{2,0}\cos^2(n_{1,0}^{1/2}gt), \tag{15b}$$

$$\langle n_3(t)\rangle = n_{2,0}\sin^2(n_{1,0}^{1/2}gt). \tag{15c}$$

b. For the chaotic pumping radiation at ω_1 it has been found that the parametric up-conversion depends essentially upon the coherence degree of pumping radiation [11].

i) For chaotic pumping radiation with extremely high coherence degree, when

$$\tau_{c,1} = \frac{1}{2\Gamma_1} \gg \frac{1}{n_{1,0}^{1/2} g} \tag{16}$$

holds, the following expressions for mean photon numbers have been found:

$$\langle n_1(t) \rangle = n_{1,0} - \frac{n_{2,0}}{2} \sin^2(2^{1/2} n_{1,0}^{1/2} gt), \tag{17a}$$

$$\langle n_2(t) \rangle = \frac{n_{2,0}}{2} [1 + \cos^2(2^{1/2} n_{1,0}^{1/2} gt)], \tag{17b}$$

$$\langle n_3(t) \rangle = \frac{n_{2,0}}{2} \sin^2(2^{1/2} n_{1,0}^{1/2} gt). \tag{17c}$$

Evidently, in this case the total energy conversion is one half of the initial photon number in the input mode, i.e. the total efficiency of the process is one half of that for coherent pumping radiation.

ii) For chaotic pumping radiation with very low coherence degree, when the condition

$$\tau_{c,1} = \frac{1}{2\Gamma_1} \ll \frac{n_{2,0}^{1/2}}{n_{1,0} g} \tag{18}$$

holds, the following simple solution for mean photon numbers have been obtained:

$$\left.\begin{aligned} \langle n_1(t) \rangle &= n_{1,0} - n_{1,0}^2 g^2 t^2 \quad &(19a)\\ \langle n_2(t) \rangle &= n_{2,0} - n_{2,0}^2 g^2 t^2 \quad &(19b)\\ \langle n_3(t) \rangle &= n_{1,0}^2 g^2 t^2 \quad &(19c) \end{aligned}\right\} \text{ for } \langle n_2(t) \rangle \geq 0, \text{ or } \langle n_3(t) \rangle \leq n_{2,0}.$$

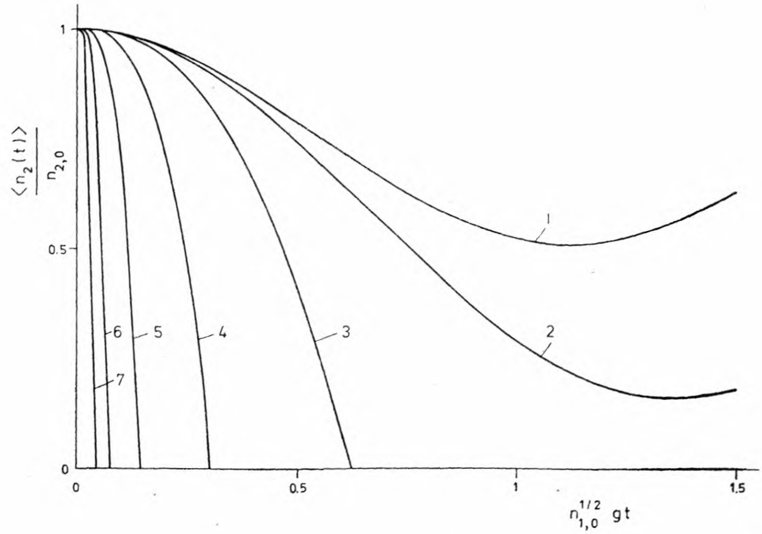
In this case the parametric up-conversion is much more effective if compared with the coherence pumping radiation.

For some special values of the typical coefficient of coherence $\eta_1 = 2\Gamma_1 / n_{1,0}^{1/2} g$ and the ratio $n_{2,0}/n_{1,0}$ the problem was solved numerically. The results are shown in the figure for $n_{2,0}/n_{1,0} = 10^{-3}$.

The curves in the figure indicate that the efficiency of parametric up-conversion increases with the decreasing coherence degree of pumping radiation.

Discussion

The influence of photon statistics of generating radiations upon the efficiency on nonlinear processes can be successfully explained by effects of intermodal correlations, as it was shown in [12, 13].



The course of the relative mean photon number in the input mode $\frac{\langle n_2(t)2\rangle}{n_{2,0}}$ at a chaotic pumping radiation for $\frac{n_{2,0}}{n_{1,0}} = 10^{-3}$ and for different values of the typical coefficient of coherence of the pumping mode $\eta_1 = \frac{2\Gamma_1}{n_{1,0}^{1/2}}$: $\eta_1 = 0\,(1)$; $\eta_1 = 10^{-3}/(2)$; $\eta_1 = 10^{-2}\,(3)$; $\eta_1 = 10^{-1}\,(4)$; $\eta_1 = 1\,(5)$; $\eta_1 = 10\,(6)$; $\eta_1 = \infty\,(7)$

Generally, the intermodal correlations between two modes of the type

$$\langle \Delta W_i(t)\, \Delta W_j(t)\rangle = \langle a_i^+(t)\, a_j^+(t)\, a_i(t)\, a_j(t) - \\ -\langle a_i^+(t)\, a_i(t)\rangle \langle a_j^+(t)\, a_j(t)\rangle \tag{20}$$

are the product of the nonlinear interaction among all three modes and they also depend strongly upon the initial statistics of generating radiations.

The positive correlation $\langle \Delta W_i \Delta W_j\rangle$ is connected with the bunching and the negative correlation $\langle \Delta W_i \Delta W_j\rangle$ with the antibunching of photons appurtenant to different modes.

The positive correlation between the subfrequency modes 1 and 2 $\langle \Delta W_1 \Delta W_2\rangle$, that is connected with bunching of photons at ω_1 and ω_2, supports the sum-frequency generation and it quenches the splitting of photons at ω_3 (differences-frequency generation). The negative correlation $\langle \Delta W_1 \Delta W_2\rangle$ supports the splitting of photons at ω_3 and it quenches the sum-frequency generation.

The correlation between any subfrequency mode i and the sum-frequency mode 3 $\langle \Delta W_i \Delta W_3\rangle (i = 1, 2)$ has a different meaning. The positive correlation $\langle \Delta W_i \Delta W_3\rangle$ supports the splitting of photons at

ω_3 and it quenches the sum-frequency generation. The negative correlation $\langle \Delta W_i \Delta W_3 \rangle$ supports the sum-frequency generation and it quenches the splitting of photons at ω_3.

In [13] it has been shown that the typical function

$$\mathscr{W}(t) = \langle \Delta W_1(t) \Delta W_3(t) \rangle + \langle \Delta W_2(t) \Delta W_3(t) \rangle - - \langle \Delta W_1(t) \Delta W_2(t) \rangle \tag{21}$$

plays a significant role in the nonlinear three mode processes.

For the positive $\mathscr{W}(t)$ the sum-frequency generation is decelerated and the splitting of photons at ω_3 (difference-frequency generation) is accelerated.

For the negative $\mathscr{W}(t)$ the sum-frequency generation is accelerated and the decay of photons at ω_3 is decelarated.

Using the conservation laws (6) we can calculate the intermodal correlations in the above description.

I. In the case of parametric generation from quentum noise the following results have been obtained:

a. For coherent pumping radiation the required intermodal correlations are as follows [12]:

$$\langle \Delta W_1(t) \Delta W_2(t) \rangle = \langle n_{1,2}(t) \rangle > 0 \tag{22a}$$

$$\langle \Delta W_1(t) \Delta W_3(t) \rangle = \langle \Delta W_2(t) \Delta W_3(t) \rangle = 0, \tag{22b}$$

and the typical function was found to be

$$\mathscr{W}(t) = -\langle n_{1,2}(t) \rangle . \tag{23}$$

The weak bunching between the subfrequency modes 1 and 2 does not affect considerably the parametric generation, provided that $n_{3,0} \gg 1$.

b. For chaotic pumping radiation the intermodal correlations were found to have the form [12]:

$$\langle \Delta W_1(t) \Delta W_2(t) \rangle \doteq n_{3,0}^2 + \langle n_3(t) \rangle^2 - -2 \langle n_3(t) \rangle n_{3,0} \exp(-2\Gamma_3 |t|), \tag{24a}$$

$$\langle \Delta W_1(t) \Delta W_3(t) \rangle = \langle \Delta W_2(t) \Delta W_3(t) \rangle = \langle n_3(t) \rangle [n_{3,0} \exp(-2\Gamma_3 |t|) - \langle n_3(t) \rangle] \tag{24b}$$

and the typical function is given by

$$\mathscr{W}(t) \doteq 4 \langle n_3(t) \rangle n_{3,0} \exp(-2\Gamma_3 |t|) - 3 \langle n_3(t) \rangle^2 - n_{3,0}^2 . \tag{25}$$

If the process were realized, i.e. the condition (12a) were satisfied, then at the beginning of the process (exactly for $\langle n_3(t) \rangle > n_{3,0}/2$) the parametric generation from quantum noise would be slightly accelerated with respect to the coherent pumping radiation. This weak acceleration would be due to the bunching between the pump mode 3 and both the subfrequency modes 1 and 2, respectively, that predominates over the bunching between the subfrequency modes 1 and 2 (see [10] and [12]).

The fact that the process is not realizable when the condition (12b) holds is due to both the bunching between the subfrequency modes 1 and 2 and the antibunching between the pumping mode 3 and the subfrequency modes 1 and 2, respectively.

II. In the case of parametric up-conversion, when regarding the conditions (14), the following approximative results have been obtained:

a. For coherent pumping radiation the intermodal correlations are given approximately by [11]

$$\langle \Delta W_1(t)\Delta W_2(t)\rangle \doteq \langle \Delta W_1(t)\Delta W_3(t)\rangle \doteq \langle \Delta W_2(t)\Delta W_3(t)\rangle = 0, \tag{26}$$

and for the typical function it holds

$$\mathscr{W}(t) \doteq 0. \tag{27}$$

In this case the nonlinear process is not considerably affected by the influence of intermodal correlations.

b. For chaotic pumping radiation the following intermodal correlations have been obtained [11]:

$$\langle \Delta W_1(t)\Delta W_2(t)\rangle \doteq -\frac{\langle n_3(t)\rangle}{2}[n_{1,0}+\langle n_1(t)\rangle], \tag{28a}$$

$$\langle \Delta W_1(t)\Delta W_3(t)\rangle = \langle n_1(t)\rangle[n_{1,0}\exp(-2\Gamma_1|t|)-\langle n_1(t)\rangle], \tag{28b}$$

$$\langle \Delta W_2(t)\Delta W_3(t)\rangle \doteq 0, \tag{28c}$$

and the typical function is given by:

$$\mathscr{W}(t) \doteq \langle n_1(t)\rangle[n_{1,0}\exp(-2\Gamma_1|t|)-\langle n_1(t)\rangle]+ + \frac{\langle n_3(t)\rangle}{2}[n_{1,0}+\langle n_1(t)\rangle]. \tag{29}$$

At an extremely high coherence degree of pumping radiation, when the relation (16) holds, the deceleration of parametric up-conversion is due to the antibunching between the modes 1 and 2 and the bunching between the modes 1 and 3.

At a very low coherence degree of pumping radiation, when the condition (18) holds, the anticorrelation between the modes 1 and 2 acts againts the sum-frequency generation, the anticorrelation between the modes 1 and 3 is, however, much more greater and causes a considerable acceleration of the process.

The above approach of treatment of the two special types of nonlinear processes is also supported by the short-time approximation solution of the problems [5, 14, 15].

A matter of special interest is the strong anticorrelation between two subfrequency modes due to parametric up-conversion with chaotic pumping light (28a), which seems to be a general property of sum-frequency generation by chaotic light [14, 15].

The formula (28a) offers an attractive experimental verification, when using the rotating ground glass disc or scattering in liquid crystals [16] for randomization of laser radiation and measuring the correlation by means of well known Hanbury Brown–Twiss intensity correlation arrangement (see e.g. [9]).

References

[1] YARIV A., *Quantum Electronics*, McGraw-Hill Book Co., New York 1964.

[2] LOUISELL W. H., *Radiation and Noise in Quantum Electronics*, McGraw-Hill Book Co., New York 1964.

[3] SMITHERS M. E., LU E. Y. C., Phys. Rev. **A10** (1974), 1874.

[4] AGRAWAL G. P., METHA C. L., J. Phys. **A7** (1974), 607.

[5] PEŘINA J., Czech. J. Phys. **B 26** (1976), 140.

[6] KIELICH S., KOZIEROVSKI M., TANAŚ R., *Antibunching in Light Harmonics Generation from Field Quantization*, 4-th Conference on Coherence and Quantum Optics, Rochester 1977.

[7] MIŠTA L., PEŘINA J., Acta Phys. Pol. **A52** (1977), 425.

[8] CROSIGNANI B., DI PORTO P., SOLIMENO S., J. Phys. **A5**, (1972), L119.

[9] PEŘINA J., *Coherence of Light*, Van Nostrand Reinhold Co., London 1972.

[10] CHMELA P., Acta Phys. Pol. **A51** (1977), 835.

[11] CHMELA P., Optical and Quant. Electr. (in press).

[12] CHMELA P., Optical and Quant. Electr. (in press).

[13] CHMELA P., Czech. J. Phys. (in press).

[14] PEŘINOVÁ V., PEŘINA J., Czech. J. Phys. **B28** (1978), 1183.

[15] PEŘINA J., *Quantum Statistical Properties of Nonlinear Optical Processes*, EKON-78, Poznań 1978 (Poland).

[16] CROSIGNANI B., DI PORTO P., BERTOLOTTI M., *Statistical Properties of Scattered Light*, Academic Press, New York 1975.

Received, September 26, 1978

Выход оптических нелинейных процессов по отношению к статистике фотонов генерирующего излучения

Обсуждено влияние статистики фотонов на протекание трёхмодовых оптических процессов. Применён метод, который основан на соблюдении статистики генерирующего излучения во время нелинейных процессов [8], а также исследованы два типичных случая, а именно, параметрическая генерация из квантового шума и параметрическая конверсия „вверх". Выяснено влияние статистики фотонов генерирующего излучения на нелинейное протекание оптических процессов с помощью эффектов межмодовой корреляции.