ATOMS, SPECTRA, RADIATION

Four-Photon Correlations upon Parametric Scattering

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Abstract—Four-photon correlations of the output radiation of a parametric amplifier with vacuum at the input are considered for an arbitrary coefficient of parametric conversion. Such states are interpreted in the literature as four-photon states. It is shown that the fourth-order correlation function for such states in the limit of a small number of photons has the asymptotics that is typical of two-photon states. Nevertheless, even in the "classical" limit of high intensities, the level of four-photon correlations, i.e., the value of the normalized fourth-order correlation function is substantially greater than that for the coherent and even thermal fields. *© 2004 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

Most of the experiments on quantum optics are related to the production of nonclassical light of different types, i.e., the light whose properties can be described only within the framework of a consistent quantum-mechanical approach. However, there exist only few types of nonclassical light that can be prepared experimentally at present. First of all, it is onephoton light, which is obtained upon one-photon transitions in atoms [1], during luminescence of quantum dots [2, 3], as well as with the help of some transformation performed with two-photon light [4]. In turn, twophoton light can be obtained upon two-photon transitions in atoms [5], but much more efficiently - due to spontaneous parametric scattering [6]. In the limit of a great number of photons, two-photon light is transformed to squeezed light, which is also nonclassical [7]. Recently two-photon light was generated due to hyper-parametric scattering [8]. Not also that both onephoton and two-photon states of light (belonging to the Fock states) are generated in all the cases mentioned above only in a superposition with the vacuum state.

The generation of other types of nonclassical light, for example, higher-order Fock states is of interest first of all from the fundamental point of view. The applications of such states are discussed in connection with the problem of quantum information [9] and the concept of quantum lithography [10]; however, these problems are far from realization at present. The attempts to prepare experimentally three- and four-photon states are mainly stimulated by the so-called Greenberger-Horne-Zeilinger (GHZ) paradox [11]. The paradox appears when one attempts to describe classically the results of the interference experiment with the state having the form

$$
|\Psi\rangle = \frac{1}{\sqrt{2}}(|++++\rangle + |---\rangle)
$$
 (1)

(the four-photon GHZ state) or

$$
|\Psi\rangle = \frac{1}{\sqrt{2}}(|+++\rangle + |---\rangle)
$$
 (2)

(the three-photon GHZ state). Here, the symbol $|+++\rangle$ denotes the state of four photons with the right circular polarization, the symbol $|- - - \rangle$ denotes the state of four photons with the left circular polarization, etc.

In [12, 13], the GHZ state was obtained from a group of four photons formed due to a random overlap of photon pairs upon parametric scattering. One of the photons serves as "trigger," while the three remaining photons form a state with the polarization part of the type (2). Groups of three photons obtained in this way were called in many papers the three-photon states. Similarly, groups of four photons appearing due to a random overlap of photon pairs are called the four-photon states [14]. It is affirmed in [14] that such groups of four photons represent the four-photon entangled states. The observation of four-photon interference was reported, i.e., the dependence of the counting rate of four-photon coincidences on the phase introduced between different groups of four photons (more exactly, between the pump pulses generating these groups). Note here that the presence of the interference pattern observed in coincidences of photocounts for four photons is well explained by the interference observed in coincidences of two photons and typical of two-photon light.

The question arises: whether the states obtained in this way can be treated as "true four-photon states"? Obviously, the answer depends on the problem for which the four-photon states are prepared. For example, the method described above is suitable for the realization the conditions of the three-photon GHZ paradox because it allows one to prepare polarization state (2).

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However, it seems that parametric scattering cannot be used to solve the problem of observation of four-photon interference [14]. Finally, of interest is the character of four-photon correlations, i.e., a set of fourth-order intensity correlation functions or, in experimental terms, the number of coincidences of photocounts for four photons. It is from this point of view that we analyze in this paper the parametric scattering of light. Because it was emphasized in [14] that parametric scattering was stimulated, we consider the case of an arbitrary coefficient of parametric amplification. Therefore, the perturbation theory, which is commonly used for the description of multiphoton correlations, proves to be inapplicable.

Three cases can be distinguished in the description of parametric scattering, which differ from each other from the point of view of photon statistics. These are the cases of the one-mode (collinear and frequencydegenerate) regime, two-mode (nondegenerate in the frequency, scattering angle or polarization), and fourmode regime (when, for example, scattering occurs at two frequency and two polarization modes). The latter regime was used in [14]. It is this regime that leads to the generation of the two-photon Bell states, i.e., the states of the type

$$
\Phi^{\pm} \equiv \frac{1}{\sqrt{2}} (|H_1 H_2 \rangle \pm |V_1 V_2 \rangle),
$$

\n
$$
\Psi^{\pm} \equiv \frac{1}{\sqrt{2}} (|H_1 V_2 \rangle \pm |V_1 H_2 \rangle).
$$
\n(3)

Here, *H* and *V* are the photon states with the horizontal and vertical polarizations, respectively, and the subscripts 1 and 2 number the frequency (or spatial) modes.

2. THE ONE-MODE REGIME

In this case, the parametric interaction Hamiltonian has the form

$$
H = \frac{1}{2} i\hbar \Gamma (a^{\dagger 2} - a^2),
$$
 (4)

where Γ is the parametric gain, a^{\dagger} and *a* are the photon creation and annihilation operators. In the first order of the perturbation theory, the state vector of the field emitted upon parametric scattering is a superposition of the vacuum and two-photon states

$$
|\psi\rangle = C_0|0\rangle + C_1|2\rangle. \tag{5}
$$

However, the exact solution is given by the vector of the state containing, except the two-photon state, also the four-photon, six-photon states, etc.:

$$
|\psi\rangle = C_0|0\rangle + C_1|2\rangle + C_2|4\rangle + C_3|6\rangle + \dots
$$

$$
= \sum_{n=0}^{\infty} C_n|2n\rangle.
$$
 (6)

It is convenient to characterize the number of groups of four photons by the fourth-order correlation function

$$
g^{(4)} = \frac{\langle a^{\dagger 4} a^4 \rangle}{\langle a^{\dagger} a \rangle^4}.
$$
 (7)

This function is measured by the number of coincidences of photocounts from four detectors in the experiment similar to the Brown–Twiss experiment (Fig. 1a), with the normalization to the product of the average detected intensities. The normalized fourth-order correlation function characterizes the radiation intensity upon the detection of four-photon effects [15]. For example, $g^{(4)} = 1$ for coherent radiation, and $g^{(4)} = 4!$ 24 for thermal (Gaussian) radiation. In the context of this paper, of interest is the value of $g^{(4)}$ for four-photon radiation (in a superposition with vacuum), which could be obtained upon the parametric decay of pump photons into groups of four photons. The state vector for such radiation has the form

$$
|\Psi\rangle = C_0|0\rangle + C_1|4\rangle.
$$

The fourth-order correlation function for this state is

$$
g^{(4)} = \frac{6}{N^3},\tag{8}
$$

where $N = \langle a^{\dagger} a \rangle$ is the average number of photons.

Consider now a state generated upon parametric scattering with the Hamiltonian (3). Within the framework of the Heisenberg approach, the correlation functions can be found exactly for any parametric gain Γ . By writing the Heisenberg equations for the creation and annihilation operators, we obtain the solution in the form

$$
a(t) = a_0 \cosh(\Gamma t) + a_0^{\dagger} \sinh(\Gamma t)
$$

and similarly for the creation operator. Here, a_0^{\dagger} and a_0 are the creation and annihilation operators at the instant $t = 0$ (or neglecting parametric interaction). The fourth-order correlation function is determined by expression (7), where $a^{\dagger} \equiv a^{\dagger}(t)$, $a \equiv a(t)$, and averaging is performed over the vacuum state. The second-order correlation function can be found similarly. As a result, we obtain

$$
g_a^4(t) = 24 + 72 \coth^3(\Gamma t) + 9 \coth^4(\Gamma t), \quad (9)
$$

$$
g_a^{(2)}(t) = 2 + \coth^2(\Gamma t). \tag{10}
$$

JOURNAL OF EXPERIMENTAL AND THEORETICAL PHYSICS Vol. 98 No. 2 2004

$$
g_a^{(4)}(t) \propto \frac{9}{(\Gamma t)^4} \propto \frac{9}{N^2}, \quad g_a^{(2)}(t) \propto \frac{1}{(\Gamma t)} \propto \frac{1}{N}
$$

when $\Gamma t \longrightarrow 0$. Such asymptotics means that only the pair correlation of photons takes place: according to (8), the four-photon states should result in a faster increase in $g^{(4)}$ at small *N*. The asymptotics at large parametric conversion coefficients $(\Gamma t \geq 1)$ gives $g_a^{(4)} \longrightarrow 105$ and $g_a^{(2)} \longrightarrow 3$. Therefore, in the limit of large gains, the output radiation of a degenerate parametric amplifier should have the super-Poisson or even super-Gaussian statistics.

The statistics of the output radiation of a degenerate parametric amplifier with vacuum at the input was studied earlier in [16], where the correlations functions of all orders were obtained in the general form, including results (9) and (10) .

3. THE TWO-MODE REGIME

Consider now parametric scattering in the nondegenerate (two-mode) case. This case is realized either upon collinear frequency-nondegenerate parametric scattering or upon noncollinear frequency-degenerate scattering or upon parametric scattering of the type II in the collinear frequency-degenerate regime. Correspondingly, photons from one pair belong to two different frequency, spatial or polarization modes. Let us denote the creation and annihilation operators in these modes by a^{\dagger} , *a* and b^{\dagger} , *b*. Then, the interaction Hamiltonian has the form

$$
H_{ab} = i\hbar\Gamma(a^{\dagger}b^{\dagger} - ab). \tag{11}
$$

By solving the Heisenberg equation, we obtain

$$
a(t) = a_0 \cosh(\Gamma t) + b_0^{\dagger} \sinh(\Gamma t),
$$

\n
$$
b(t) = b_0 \cosh(\Gamma t) + a_0^{\dagger} \sinh(\Gamma t).
$$
\n(12)

The exact expressions for the fourth- and second-order correlation functions are

$$
g_{ab}^{(4)}(t) \equiv \frac{\langle (a^{\dagger})^2 (b^{\dagger})^2 a^2 b^2 \rangle}{\langle a^{\dagger} a \rangle^2 \langle b^{\dagger} b \rangle^2}
$$
(13)

 $= 4 + 16 \coth^2(\Gamma t) + 4 \coth^4(\Gamma t),$

$$
g_{ab}^{(2)}(t) = \frac{\langle a^{\dagger} b^{\dagger} a b \rangle}{\langle a^{\dagger} a \rangle \langle b^{\dagger} b \rangle} = 1 + \coth^2(\Gamma t) \qquad (14)
$$

JOURNAL OF EXPERIMENTAL AND THEORETICAL PHYSICS Vol. 98 No. 2 2004

Fig. 1. Scheme of the experiment for measuring $g^{(4)}$ for the output radiation of a parametric converter. Only pump radiation (p) is incident on a nonlinear crystal; the signal (s) and idle (2I) modes correspond to vacuum. (a) The one-mode regime: the signal and idle photons belong to one spatial and one frequency mode. The fourth-order correlation function is measured using three nonpolarizing beamsplitters BS1, BS2, and BS3, four photodetectors, and the fourfold photocount coincidence scheme CC. (b) The two-mode regime: the signal and idle photons belong to different modes (in this case, different spatial modes). The fourthorder correlation function is measured using two nonpolarizing beamsplitters BS1 and BS2, four photodetectors, and the fourfold photocount coincidence scheme CC. (c) The regime of generation of the Bell states. The signal and idle photons are emirred to two spatial and two polarization modes. The fourth-order correlation function is measured using two nonpolarizing beamsplitters BS1 and BS2, four photodetectors, and the fourfold photocount coincidence scheme CC

(the scheme for measuring $g_{ab}^{(4)}$ is shown in Fig. 1b). One can see that the asymptotics of correlation functions at small parametric gains is also has a "two-photon" character, i.e., $g_{ab}^{(4)}$ has the order $1/N^2$ and $g_{ab}^{(4)}(t)$ has the order 1/*N*. For large parametric gains, we obtain $g_{ab}^{(4)} \longrightarrow 24$ and $g_{ab}^{(2)} \longrightarrow 2$. Such statistics could be

1

1

2 1

inherent in thermal radiation, but we should take into account that in this case two different modes *a* and *b* are involved. Therefore, the statistics can be called super-Gaussian in the two-mode case as well.

4. GENERATION OF THE BELL STATES

Consider now the regime of parametric scattering that was used in [14] and leads, in the limit of small parametric conversion coefficient, to the generation of one of the Bell states (3), namely, the state Ψ ⁻ in a superposition with vacuum. As above, we consider the case of an arbitrary parametric conversion coefficient. The interaction Hamiltonian has the form

$$
H_{a_H b_V} = i\hbar \Gamma (a_H^{\dagger} b_V^{\dagger} - a_V^{\dagger} b_H^{\dagger}) + \text{H.c.}
$$
 (15)

Here, a^{\dagger} and b^{\dagger} are, as before, the creation and annihilation operators for the two modes, which can be frequency or spatial modes, and the subscripts *H* and *V* denote vertical and horizontal polarizations, respectively.

The solution for the creation and annihilation operators has the form

$$
a_H(t) = a_{H0} \cosh(\Gamma t) + b_{V0}^{\dagger} \sinh(\Gamma t),
$$

\n
$$
b_V(t) = b_{V0} \cosh(\Gamma t) - a_{H0}^{\dagger} \sinh(\Gamma t),
$$

\n
$$
a_V(t) = a_{V0} \cosh(\Gamma t) - b_{H0}^{\dagger} \sinh(\Gamma t),
$$
\n(16)

$$
b_H(t) = b_{H0} \cosh(\Gamma t) + a_{V0}^{\dagger} \sinh(\Gamma t).
$$

For the fourth- and second-order correlation functions, we obtain

$$
g_{a_{H}b_{V}a_{V}b_{H}}^{(4)}(t)
$$
\n
$$
= \frac{\langle a_{H}^{\dagger}b_{V}^{\dagger}a_{V}^{\dagger}b_{H}^{\dagger}a_{H}b_{V}a_{V}b_{H} \rangle}{\langle a_{H}^{\dagger}a_{H} \rangle \langle b_{V}^{\dagger}b_{V} \rangle \langle a_{V}^{\dagger}a_{V} \rangle \langle b_{H}^{\dagger}b_{H} \rangle}
$$
\n
$$
= 1 + 2 \coth^{2}(\Gamma t) + \coth^{4}(\Gamma t), \qquad (17)
$$

$$
g_{a_{H}b_{V}}(t) = \frac{\langle a_{H}^{\dagger}b_{V}^{\dagger}a_{H}b_{V}\rangle}{\langle a_{H}^{\dagger}a_{H}\rangle\langle b_{V}^{\dagger}b_{V}\rangle} = 1 + \coth^{2}(\Gamma t).
$$

One can see that statistics in this case is characterized by even lower value of four-photon correlations than in the two-mode case, but, nevertheless, is super-Gaussian: $g_{a_{H}b_{V}}^{(4)} \longrightarrow 4$ and $g_{a_{H}b_{V}}^{(2)} \longrightarrow 2$ for $\Gamma t \longrightarrow 0$. The scheme for measuring the corresponding fourth-order correlation function is shown in Fig. 1c. We can also consider the case when the fourth-order moment is measured in the same regime of parametric scattering (Fig. 1c):

$$
g_{a_\mu b_\nu a_\mu b_\nu}^{(4)}(t)\,=\,\frac{\big\langle a_H^{\dagger 2}b_V^{\dagger 2}a_H^2b_H^2\big\rangle}{\big\langle a_H^{\dagger}a_H\big\rangle^2\big\langle b_V^{\dagger}b_V\big\rangle^2}.
$$

This correlation function is equal to $4 + 16 \coth^2(\Gamma t) +$ $4 \coth^4(\Gamma t)$, as in the two-mode case.

Therefore, the state generated upon parametric scattering is characterized by substantially weaker fourphoton correlations than the "true four-photon state", which could be obtained, for example, due to the decay of pump photons into groups of four photons in a medium with the fourth-order nonlinearity (of course, the probability of such process is extremely low). Nevertheless, four-photon correlations for this state even in the limit of large parametric gains are substantially stronger than those for classical sources with the Poisson or Gaussian statistics. In this sense, the strongest correlations should be observed in the degenerate regime of parametric scattering.

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SPELL: 1. beamsplitters, 2. emirred