Spectral properties of three-photon entangled states generated via three-photon parametric down-conversion in a $\chi^{(3)}$ medium

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We consider the quantum state of light produced via direct parametric decay of pump photons into photon triples in a medium with cubic nonlinearity. For this state generated in the near-collinear frequency-degenerate regime, the third- and second-order Glauber's correlation functions are calculated and the intensity distribution over frequency and wave vector is found. It is shown that the number of photons generated into a single mode via the three-photon down-conversion is proportional to the width of the frequency-angular intensity distribution for the corresponding two-photon phase matching (spontaneous parametric down-conversion). The intensity of three-photon parametric down-conversion is shown to have an extremely broad frequency spectrum, even for a fixed angle of scattering.

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I. INTRODUCTION

The generation of nonclassical states of light is one of the main directions in quantum optics. At the same time, the number of such states available at present is very limited. Within the class of photon-number (Fock) states, only two types of light can be generated in practice: one-photon light and two-photon light. Several attempts have been made to produce higher-order Fock states via the process providing two-photon light: spontaneous parametric down-conversion (SPDC) [1]. Light generated via SPDC contains only even numbers of photons [2]: mostly, pairs of photons, but also four-photon groups, six-photon groups, etc. Due to the existence of photon quadruples in this radiation, it was possible to use it for the production of three-photon polarization Greenberger-Horne-Zeilinger (GHZ) states [3]. However, while the polarization part of the state prepared from SPDC radiation has the required GHZ form, the photon statistics is still typical for two-photon light. Indeed, from the asymptotic behavior of the four-photon Glauber's correlation function, one can see [4] that the number of photons as well as photon pairs in SPDC radiation scales linearly in the pump intensity while the number of photon quadruples scales quadratically. For a four-photon Fock state generated in superposition with vacuum, the number of photon quadruples should be linear in the photon number. In the case of pulsed SPDC, there is only a twofold increase in the normalized four-photon correlation function, and hence, the state remains qualitatively the same as for cw SPDC [5].

Direct generation of the Fock states of order higher than 2 is quite challenging. For instance, if one considers the generation of N-photon states via N-photon parametric decay of pump photons in a nonlinear crystal, then the efficiency is proportional to the squared Nth-order nonlinear susceptibility. Usually, the $N+$first-order nonlinear susceptibility is smaller than the Nth-order one by a value on the order of the interatomic field (about five orders of magnitude in esu units), so even the three-photon parametric down-conversion is very difficult to observe.

A possible way to generate three-photon entangled states was discussed in Ref. [6], where it was suggested to produce the third photon by up-converting two idler photons of an entangled pair. Unfortunately, no experimental evidence for this effect has been obtained until now. Probably, this is because of the small efficiency of this process, which is determined by the sixth power of the quadratic susceptibility and the squared intensity of the pump.

In this paper we consider in detail three-photon parametric down-conversion. This process has been much considered in the literature in connection with so-called generalized squeezing (see, for instance, [7–9])—i.e., squeezing originating from not a quadratic but a higher-order interaction Hamiltonian. However, no third-order squeezing has been observed in experiment so far, again because of the extremely small value of the third-order optical nonlinearity. As a more feasible scheme for observing three-photon down-conversion and the corresponding squeezing, an intracavity version was proposed in [10]. The research on the squeezing properties of three-photon down-conversion was naturally focused on the continuous variables, such as quadratures and their noise, as well as various quasiprobability functions like the Wigner function or the Glauber-Sudarshan function.

In the present paper, we focus not on the continuous-variable properties of the three-photon parametric down-conversion but on its photon statistics. On the one hand, this means that the regime under study will be a spontaneous one, in which the generated fields are weak while the pump is strong, nondepleted, and described classically. This regime should result in the generation of three-photon entangled states rather than three-photon squeezed states. On the other hand, the values of interest here will be the photon-number moments of various order: intensity and the second-order and third-order Glauber's correlation functions.

The paper is organized as follows. In Sec. II, we discuss the main feature of the phase matching conditions for the

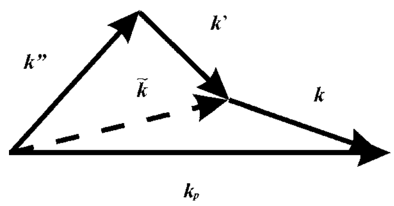

FIG. 1. Phase matching for the three-photon PDC: due to the "loose" phase matching condition, the process "visualizes" zero-point vacuum fluctuations integrated over all possible frequencies and wave vectors for the corresponding two-photon process.

three-photon parametric down-conversion (PDC): namely, the fact that the number of constraints is less than the number of "free parameters." This leads to the main idea of the paper: namely, that the efficiency of three-photon PDC is determined not by the brightness of zero-point vacuum fluctuations, as the efficiency of the two-photon PDC, but by the integral of this brightness over all modes involved. In the next sections, the three-photon quantum state is calculated (Sec. III) and the correlation functions and intensity spectrum for this state are found (Sec. IV). Finally, in Sec. V, we discuss the applicability of the collinear frequency-degenerate approximation used in the previous sections and extend the approach to a more general case.

II. THREE-PHOTON PARAMETRIC DOWN-CONVERSION AND THE ZERO-POINT VACUUM FLUCTUATIONS

The most important feature distinguishing the three-photon PDC from the well-known two-photon PDC "SPDC" is a more "loose" phase matching. Indeed, for SPDC, there are two participating modes (two photons), each described by three free parameters, and two constraints, one of them of scalar form (frequency phase matching condition) and the other of vector form (wave vector phase matching condition). For the three-photon PDC, there is one more mode involved but the number of constraints is the same. As a result, if one is interested in the efficiency of the three-photon PDC, in which one of the three photons is scattered into a given mode $\mathbf{k}$, ω (Fig. 1), there are still infinitely many ways to satisfy the phase matching conditions.

This can be demonstrated by introducing the frequency $\tilde{\omega}\equiv\omega_p-\omega$ and the wave vector $\tilde{\mathbf{k}}\equiv\mathbf{k}_p-\mathbf{k}$, where ω_p and $\mathbf{k}_p$ are the pump frequency and wave vector for the three-photon PDC. Then, the possibility of having one of the photons scattered into a mode $\mathbf{k}$, ω corresponds to all possible two-photon PDC processes that can be realized with the pump having the wave vector $\tilde{\mathbf{k}}$ and the frequency $\tilde{\omega}$ (shown in Fig. 1). In this connection, it is useful to employ the concept of zero-point vacuum fluctuations, which is very helpful in interpreting the spectra and calculating the efficiency of SPDC [1]. In the case of SPDC, when one looks for the number of photons (say, signal photons) scattered into a given mode, the conjugated idler mode is found in a unique way. Hence, the number of signal photons is proportional to the number of photons in the idler mode, the so-called "brightness of the vacuum" [1]. One can say that the intensity of two-photon PDC at some frequency and wave vector is "seeded" by zero-point vacuum fluctuations at the conjugate frequency and wave vector.

The same reasoning, in the case of the three-photon PDC, leads us to the conclusion that the number of photons scattered into the mode $\mathbf{k}$, ω via a three-photon PDC is proportional to the product of the "brightnesses of the vacuum" for the two conjugate modes integrated over all possible frequencies and wave vectors for one of them. In other words, the intensity of three-photon PDC at a certain frequency ω and wave vector $\mathbf{k}$ is "seeded" by zero-point vacuum fluctuations at a continuum of frequencies and wave vectors, corresponding to all possible ways for two-photon PDC with pump frequency $\tilde{\omega}$ and wave vector $\tilde{\mathbf{k}}$. Hence, unlike in the case of SPDC, where the spectral width of phase matching matters only for the integral intensity of the scattered photons, in the case of the three-photon PDC it also matters for the "differential" intensity. This means that by choosing a medium with broad frequency-angular spectrum (for the corresponding two-photon process) one can considerably enhance the efficiency of the three-photon PDC.

These rather qualitative considerations will be further developed in the next section.

III. THREE-PHOTON STATE VECTOR

Consider the three-photon PDC in a medium with cubic susceptibility $\chi^{(3)}$. In our consideration we will follow the approach used in the calculation of the intensity and correlation functions for two-photon light [1,11,12]. In the dipole approximation, the interaction Hamiltonian has the form

$$\mathcal{H}=\int \mathrm{d}^3\mathbf{r}\chi^{(3)}(\mathbf{r})E^4(\mathbf{r},t), \qquad (1)$$

where E is the electric field and the integral runs over the volume of the crystal where PDC takes place. One of the field modes is occupied by the pump, which is cw laser radiation and hence well described by the classical analytic signal $E_0^{(+)}(\mathbf{r},t)=E_0\exp(-i\omega_p t+i\mathbf{k}_p\cdot\mathbf{r})$. In the other modes, there is vacuum initially, and hence, the field in these modes requires a quantum description. The corresponding field operators can be expressed in terms of photon creation and annihilation operators:

$$E^{(-)}(\mathbf{r},t)=-\frac{i}{v}\int \mathrm{d}^3\mathbf{k}c_{\mathbf{k}}a^{\dagger}(\mathbf{k})\exp[i\omega(\mathbf{k})t-i\mathbf{k}\cdot\mathbf{r}], \qquad (2)$$

where

$$c_{\mathbf{k}}\equiv\sqrt{\frac{\hbar v\,\omega(\mathbf{k})u(\omega)}{(2\pi)^2 cn(\omega)}}, \qquad (3)$$

$u(\omega)$ is the group velocity, v the quantization volume in $\mathbf{k}$ space, c the speed of light, and $n(\omega)$ the refractive index.

Then the Hamiltonian can be written in the form

$$\mathcal{H}=\frac{i}{v^3}E_0\int \mathrm{d}^3\mathbf{r}\chi^{(3)}(\mathbf{r})\int\int\int \mathrm{d}^3\mathbf{k}\mathrm{d}^3\mathbf{k}'\mathrm{d}^3\mathbf{k}''$$

$$\times c_{\mathbf{k}}c_{\mathbf{k}'}c_{\mathbf{k}''}a^{\dagger}(\mathbf{k})a^{\dagger}(\mathbf{k}')a^{\dagger}(\mathbf{k}'')\exp(i\Delta\mathbf{k}\cdot\mathbf{r}-i\Delta\omega t)+\text{H.c.},$$

$$\Delta\mathbf{k} \equiv \mathbf{k}(\omega) + \mathbf{k}'(\omega') + \mathbf{k}''(\omega'') - \mathbf{k}_p,$$

$$\Delta\omega \equiv \omega + \omega' + \omega'' - \omega_p, \tag{4}$$

where $a^\dagger(\mathbf{k})$, $a^\dagger(\mathbf{k}')$, and $a^\dagger(\mathbf{k}'')$ are photon creation operators in three field modes with wave vectors $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{k}''$ and frequencies ω, ω', and ω'', respectively. It is supposed that the nonlinear medium is transparent and its dispersion is given by the dependence $k(\omega)$.

Suppose that the three-photon PDC occurs in a crystal with length L along the z direction, the pump is a Gaussian beam with diameter a, and its wave vector $\mathbf{k}_p$ is directed along z. Let us denote by k, k', and k'' the absolute values of the wave vectors, by k_z, k_z', and k_z'' their longitudinal components, and by $\mathbf{q}$, $\mathbf{q}'$, and $\mathbf{q}''$ their transverse components. From the dispersion relations, we have $k=n(\omega)\omega/c$ and similarly for k' and k''. In the expressions for $c_{\mathbf{k}}$, the group velocity can be assumed approximately equal to the phase velocity. The three waves corresponding to the generated photons are supposed to be "ordinary," while the pump wave "extraordinary" (one can call it "type-I" or "e-ooo" phase matching, similarly to the two-photon case). The distribution of $\chi^{(3)}$ over the crystal is assumed to be uniform; hence the integral over spatial coordinates will be bounded by $\pm L/2$ in the longitudinal direction and by the factor $e^{-(x^2+y^2)/a^2}$ in the transverse directions. It is convenient to denote the vector $\{x;y\}$ as ρ.

The state vector calculated to the first order of the perturbation theory will contain an integral of $\mathcal{H}$ over time; this leads to the appearance of the delta function $\delta(\Delta\omega)$. This delta function is usually interpreted as the frequency phase matching, or stationarity, condition $\omega+\omega'+\omega''-\omega_p=0$.

Following [12], we will introduce the transverse mismatch $\Delta\mathbf{q} \equiv \mathbf{q}+\mathbf{q}'+\mathbf{q}''$ and the longitudinal mismatch $\Delta k_z \equiv k_p - k_z - k_z' - k_z''$. Let the process be near collinear—i.e., $q \ll k, q' \ll k', q'' \ll k''$. Then the generated state can be written as

$$|\psi\rangle = |\text{vac}\rangle + \frac{\hbar^{1/2}\chi^{(3)}E_0}{(2\pi)^2 c^3 v^{3/2}} \int\int\int d^2\mathbf{q}\,d^2\mathbf{q}'\,d^2\mathbf{q}'' \int\int d\omega\, d\omega' \times \sqrt{\omega\omega'(\omega_p-\omega-\omega')}F_z(\Delta k_z)F_\rho(\Delta\mathbf{q}) \times a^\dagger(\omega,\mathbf{q})a^\dagger(\omega',\mathbf{q}')a^\dagger(\omega_p-\omega-\omega',\mathbf{q}'')|\text{vac}\rangle, \tag{5}$$

where

$$F_z(\Delta k_z) \equiv \int_{-L/2}^{L/2} dz\, e^{i\Delta k_z z},$$

$$F_\rho(\Delta\mathbf{q}) \equiv \int d^2\rho\, e^{i\Delta\mathbf{q}\rho} e^{-\rho^2/a^2}. \tag{6}$$

Now, let us also accept the approximation of near-degenerate phase matching, with the central frequencies of the three photons denoted as $\omega_0 \equiv \omega_p/3$. Then, after substituting $\omega \equiv \omega_0 + \Omega$ and $\omega' \equiv \omega_0 + \Omega'$ and expanding the longitudinal wave vector components to the second order in Ω and Ω' and in q, q', and q'', we obtain the longitudinal mismatch in the form

$$\Delta k_z = \frac{1}{2k_0}[q^2 + q'^2 + q''^2] - \frac{1}{2s}[\Omega^2 + \Omega'^2 + (\Omega+\Omega')^2], \tag{7}$$

where $k_0 \equiv k(\omega_0)$ and s is inversely proportional to the group velocity dispersion of the nonlinear medium: $s \equiv (d^2k/d\omega^2)^{-1}$.

With these assumptions, the expression for the state vector of the generated light (whose three-photon part can be called the "triphoton" [6]) becomes

$$|\psi\rangle = |\text{vac}\rangle + \frac{\hbar^{1/2}\chi^{(3)}E_0\omega_0^{3/2}a^2L}{4\pi c^3 v^{3/2}} \times \int\int\int d^2\mathbf{q}\,d^2\mathbf{q}'\,d^2\mathbf{q}'' \int\int d\Omega\, d\Omega'\, e^{-a^2(\mathbf{q}+\mathbf{q}'+\mathbf{q}'')^2/4} \times \text{sinc}\,\frac{L}{4}\left(\frac{q^2+q'^2+q''^2}{k_0} - \frac{\Omega^2+\Omega'^2+(\Omega+\Omega')^2}{s}\right) \times a^\dagger(\omega_0+\Omega,\mathbf{q})a^\dagger(\omega_0+\Omega',\mathbf{q}')a^\dagger(\omega_0-\Omega-\Omega',\mathbf{q}'') \times|\text{vac}\rangle, \tag{8}$$

where $\text{sinc}(x) \equiv \sin(x)/x$.

Further, one can note that the integration in q'' can be eliminated in two cases: when the exponential factor in the integrand is much narrower in q'' than the "sinc" function (the case of a short crystal) and the opposite case, when the crystal is very long. For typical parameters $L \simeq 5$ cm and $a = 0.1$ cm, the first situation is the case, and hence the integral can be replaced by $1/\pi a^2$, with $\mathbf{q}''$ fixed at $-(\mathbf{q}+\mathbf{q}')$.

As a result, the final expression for the state vector is

$$|\psi\rangle = |\text{vac}\rangle + \frac{\hbar^{1/2}\chi^{(3)}E_0\omega_0^{3/2}L}{(2\pi)^2 c^3 v^{3/2}} \int\int d^2\mathbf{q}\,d^2\mathbf{q}' \int\int d\Omega\, d\Omega' \times \text{sinc}\,\frac{L}{4}\left(\frac{q^2+q'^2+(\mathbf{q}+\mathbf{q}')^2}{k_0} - \frac{\Omega^2+\Omega'^2+(\Omega+\Omega')^2}{s}\right) \times a^\dagger(\omega_0+\Omega,\mathbf{q})a^\dagger(\omega_0+\Omega',\mathbf{q}')a^\dagger(\omega_0-\Omega-\Omega', -(\mathbf{q}+\mathbf{q}'))|\text{vac}\rangle. \tag{9}$$

IV. CALCULATION OF CORRELATION FUNCTIONS AND INTENSITY DISTRIBUTION

Now, having obtained the expression (9) for the three-photon state, let us calculate some quantities measurable in experiment. These will be the third-order intensity correlation function, the second-order intensity correlation function, and the intensity distribution.

A. Third-order correlation function

In the calculation of the third-order correlation function, it is reasonable to consider the case where triple coincidences are registered for the radiation emitted collinearly to the pump beam [Fig. 2(a)], within the whole spectral band al-

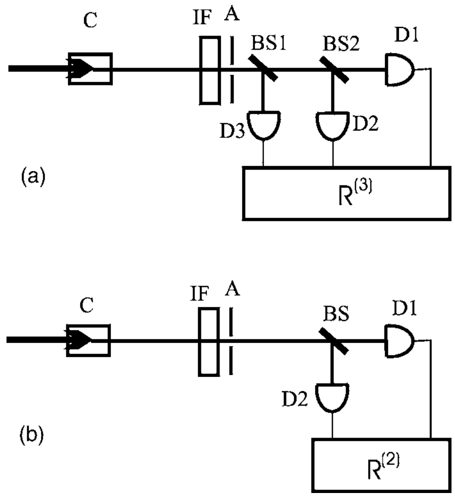

FIG. 2. Setups for the measurement of the third-order Glauber's correlation function (a) and the second-order Glauber's correlation function (b). For the three-photon radiation generated in the crystal *C*, collinear direction is selected by aperture *A*. The interference filter IF serves for parasite light suppression. BS are beam splitters and *D*1, *D*2, and *D*3 photon-counting detectors.

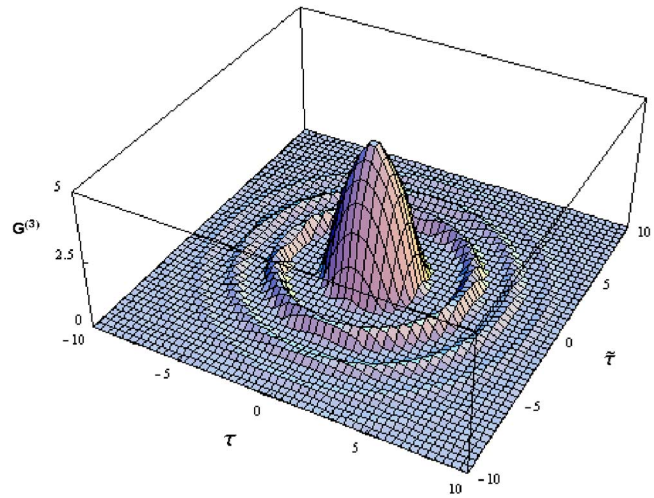

FIG. 3. (Color online) A typical shape of the third-order correlation function for the three-photon light.

lowed by the phase matching (this spectral band will be calculated in the subsection devoted to the intensity distribution). Then, the registration scheme should include an aperture for the transverse mode selection, an interference filter for selecting the spectral band corresponding to phase matching, a 33%-67% beam splitter, a 50%-50% beam splitter, and three detectors connected to a triple-coincidence scheme. Suppose, as the first step, that the system has an infinitely high time resolution, so that one can measure the probability that the first detector clicks at a time t, the second detector at a time $t+\tau$, and the third detector at a time $t+\tilde{\tau}$. This probability is defined by the third-order Glauber's correlation function [13]

$$G^{(3)}(t,\tau,\tilde{\tau}) = \| \Phi^{(3)}\rangle|^2, \tag{10}$$

$$|\Phi^{(3)}\rangle \equiv E_1^{(+)}(t)E_2^{(+)}(t+\tau)E_3^{(+)}(t+\tilde{\tau})|\psi\rangle, \tag{11}$$

where $E_1^{(+)}$, $E_2^{(+)}$, and $E_3^{(+)}$ are positive-frequency field operators on the detectors, and in the expressions for these fields [similar to Eq. (2)], the photon annihilation operators are supposed to relate to the modes with zero transverse wave vector—for instance,

$$E_1^{(+)} \propto \int \mathrm{d}\omega_1 a(\omega_1, \mathbf{q}_1 = 0)\exp(i\omega_1 t). \tag{12}$$

Calculation of the vector $|\Phi^{(3)}\rangle$ from Eq. (11) leads to the disappearance of the integrals over the transverse wave vectors in expression (9) for ψ. When doing the integrals in Ω, Ω', it is convenient to pass to the new frequency variables

$$\Omega_\pm \equiv \frac{\Omega \pm \Omega'}{\sqrt{2}}. \tag{13}$$

Then, after rather bulky but simple algebra, the result is

$$G^{(3)}(\tau,\tilde{\tau}) \propto [\chi^{(3)}]^2 I_p s^2 F^2\left(\sqrt{\frac{2s}{3L}}(\tau+\tilde{\tau}), \sqrt{\frac{2s}{L}}(\tau-\tilde{\tau}) \right), \tag{14}$$

where I_p is the pump intensity and the function F is defined as

$$F(x,x') \equiv \pi \int_0^\infty \mathrm{d}\kappa J_0[\sqrt{\kappa(x^2+x'^2)}]\mathrm{sinc}(\kappa), \tag{15}$$

J_0 being the zeroth-order Bessel function. Note that the argument t of the correlation function disappears due to the stationarity of the field.

The third-order correlation function $G^{(3)}$ or the number of triple coincidences registered by a setup shown in Fig. 2(a), as we see from Eqs. (14) and (15), is symmetric with respect to its time arguments or to the delays between the "clicks" of any two detectors. (One can show that if we introduce the third time delay $\tau' \equiv \tau - \tilde{\tau}$ in the coordinates τ and $\tilde{\tau}$, $G^{(3)}$ will have the same shape.) The typical shape of $G^{(3)}(\tau,\tilde{\tau})$ is shown in Fig. 3(a).

The width of the correlation function $G^{(3)}(\tau,\tilde{\tau})$ in both coordinates is $\sqrt{3L/8s}$. As one could expect, it gets broader in a long crystal with large GVD. For typical values of L on the order of several centimeters and s on the order of 10^{27} cm/s^2, the width of the correlation function is on the order of tens of femtoseconds, as in the case of SPDC [14].

Since in a real experimental setup time resolution is always worse than that, it is reasonable to calculate the total counting rate of triple coincidences by integrating $G^{(3)}$ over its both arguments. The resulting value, whose physical meaning is the number of coincidences registered with a low-resolution coincidence circuit, is

$$R^{(3)} \propto \int\int \mathrm{d}\tau \mathrm{d}\tilde{\tau} G^{(3)}(\tau,\tilde{\tau}) \propto [\chi^{(3)}]^2 I_p s L. \tag{16}$$

Hence, the total number of triple coincidences scales as the product of the pump power, the squared cubic susceptibility, the length of the nonlinear medium, and the inverse group velocity dispersion.

B. Second-order correlation function

Our calculation of the second-order correlation function will also relate to the case where the registered radiation is emitted collinearly to the pump beam [Fig. 2(b)]. This time, the setup includes only two detectors, a 50% beam splitter and a pair coincidence circuit. The correlation function is calculated as

$$G^{(2)}(t,\tau) = \| |\Phi^{(2)}\rangle|^2, \tag{17}$$

where

$$|\Phi^{(2)}\rangle \equiv E_1^{(+)}(t)E_2^{(+)}(t+\tau)|\psi\rangle \tag{18}$$

and $E_1^{(+)}$ and $E_2^{(+)}$ are positive-frequency field operators on the detectors. Like in the previous subsection, the corresponding photon annihilation operators are supposed to relate to the modes with zero transverse wave vector. Because of this, the integrals in the transverse wave vectors vanish.

Passing, as in the previous case, to the sum and difference frequencies (13), we obtain the second-order correlation function in the form

$$G^{(2)}(\tau) \propto [\chi^{(3)}]^2 I_p s^{3/2} L^{1/2} \int \mathrm{d}x F^2\left(\sqrt{\frac{2s}{L}}\tau, x\right), \tag{19}$$

with the function F defined in Eq. (15).

From Eqs. (19) and (14) one can see that the shape of the second-order correlation function for three-photon light is given by the integral of the third-order correlation function with respect to one of its two time arguments. The typical width of the second-order correlation function is $\sqrt{L/2s}$. Integrating the correlation function over the time delay τ, we obtain the counting rate of pair coincidences,

$$R^{(2)} \propto \int\int \mathrm{d}\tau G^{(2)}(\tau) \propto [\chi^{(3)}]^2 I_p s L. \tag{20}$$

The numbers of triple and pair coincidences, which differ by only a numerical factor, both scale as the product of the pump intensity, the crystal length, the squared cubic susceptibility, and the inverse group velocity dispersion. This agrees with the requirement that the numbers of "photon triples" and "photon pairs" in three-photon light should both scale as the number of photons.

C. Intensity distribution

The number of photons emitted in three-photon PDC will be the subject of the present section. In this case, in contrast to the way we calculated the correlation functions, we will study not only the number of photons scattered in the direction exactly collinear to the pump and into the whole spectral band. Instead, we will consider the frequency–wave-vector spectrum of the scattered photons, or intensity, as a function of the transverse wave vector $\mathbf{q}_1$ and the detuning $\Omega_1 \equiv \omega_1 - \omega_0$ of the observation frequency ω_1 from degenerate phase matching. This intensity distribution can be measured by a setup with frequency- and angular-selective detection (like the setups used for obtaining frequency-angular spectra of SPDC; see [15,16]) and can be calculated as

$$I(\omega_1, q_1) = \| |\Phi\rangle|^2, \tag{21}$$

where

$$|\Phi\rangle \equiv E^{(+)}(t,\rho)|\psi\rangle,$$

$$E^{(+)}(t,\rho) = ic_{\mathbf{k}_1} a(\mathbf{k}_1)\exp(i\omega_1 t - i\mathbf{q}_1\rho),$$

the wave vector $\mathbf{k}_1$ having the transverse component $\mathbf{q}_1$ and length $k(\omega_1)$.

After substituting Eq. (9) for $|\psi\rangle$, the calculation runs as in the previous cases. Due to the photon annihilation operator $a(\mathbf{k}_1)$, the integrals in Ω' and $\mathbf{q}'$ in the expression for $|\Phi\rangle$ vanish. Then, after taking the squared norm of $|\Phi\rangle$, we obtain the intensity distribution in the form

$$I(\Omega_1, q_1) \propto [\chi^{(3)}]^2 I_p L^2 \int\int \mathrm{d}\Omega \mathrm{d}^2\mathbf{q} \times \mathrm{sinc}^2 \frac{L}{4}\left[\frac{q^2 + q_1^2 + (\mathbf{q}+\mathbf{q}_1)^2}{k_0} - \frac{\Omega^2 + \Omega_1^2 + (\Omega+\Omega_1)^2}{s}\right]. \tag{22}$$

In the maximum, which corresponds to exactly collinear frequency-degenerate phase matching, the intensity is

$$I(0,0) \propto [\chi^{(3)}]^2 I_p L^2 \int\int \mathrm{d}\Omega \mathrm{d}^2\mathbf{q} \times \mathrm{sinc}^2 \frac{L}{2}\left[\frac{q^2}{k_0} - \frac{\Omega^2}{s}\right]. \tag{23}$$

It is interesting to calculate the maximal number of photons per mode generated via three-photon PDC. Taking into account all factors in the expression for the intensity and passing to the number of photons per mode, we find that the expression is similar to the well-known Klyshko result for SPDC (see, for instance, [1]),

$$N = \frac{(2\pi)^2 [\chi^{(2)}]^2 E_0^2 \omega_0^2 L^2}{c^2}, \tag{24}$$

but with the squared quadratic susceptibility $[\chi^{(2)}]^2$ replaced by the product of the squared cubic susceptibility and the squared effective "vacuum field:"

$$[\chi^{(2)}]^2 \to [\chi^{(3)}]^2 E_{\mathrm{vac}}^2. \tag{25}$$

The square of the "effective vacuum field" can be found from the expression for the vacuum brightness [1],

$$E_{\mathrm{vac}}^2 = \frac{2\pi\hbar\omega_0}{c}\Delta^2\mathbf{q}\Delta\Omega, \tag{26}$$

and $\Delta^2\mathbf{q}\Delta\Omega \equiv S$ can be defined as the area in $\mathbf{q},\Omega$ space where the "sinc" function under the integral in Eq. (23) is essentially nonzero. Alternatively, S can be called the integral spectrum of zero-point vacuum fluctuations "seeding" the three-photon PDC.

Calculation of $S=\Delta^2\mathbf{q}\Delta\Omega$ deserves a separate discussion. Calculating it in the near-collinear near-degenerate approximation, which was used above, is not quite correct. Indeed, the frequency–wave-vector domain where the integrand of Eq. (23) is nonzero actually coincides with the spectrum of SPDC that would be obtained from a pump with the frequency $\tilde{\omega}$ and wave vector $\tilde{\mathbf{k}}$ (see Fig. 1), and the SPDC spectrum is much broader than its collinear frequency-degenerate part. The range of two-photon decay phase matching extends in frequency over all visible and IR ranges and in angles over tens of degrees. Therefore, the calculation should be performed without expanding the longitudinal mismatch, and the result will depend not only on k_0 and s, but on the whole dispersion dependence.

V. MORE GENERAL APPROACH

To understand the validity of the near-collinear near-degenerate approximation used above, one should pass to a more general approach, which means avoiding the expansion (7). As a result, the state vector (9) will contain an exact expression for the longitudinal mismatch $\Delta k_z(\Omega,\mathbf{q})$ in the argument of the "sinc" function. Also, it will be no more possible to move the frequency factors from under the integral in Eq. (5). This will lead to the appearance of the factor $f(\Omega,\Omega')\equiv\sqrt{(\omega_0+\Omega)(\omega_0+\Omega')(\omega_0-\Omega-\Omega')}$ under the integral of (9).

Then, the intensity distribution (22) will become

$$I(\Omega_1,q_1)\propto[\chi^{(3)}]^2 I_p L^2 \times\int\int d\Omega d^2\mathbf{q} f^2(\Omega,\Omega_1)\mathrm{sinc}^2\frac{L}{2}\Delta k_z(\Omega,\mathbf{q}), \tag{27}$$

where $\Delta k_z(\Omega,\mathbf{q})$ is calculated directly from the frequencies, angles, and the dispersion law.

As an example, in Fig. 4 we present the frequency-angular spectrum of two-photon PDC calculated as the integrand of Eq. (27) for the case of lithium formate monohydrate ($HCOOLi\cdot H_2O$) crystal, the pump having wavelength $\tilde{\lambda}=526.5$ nm and its wave vector $\tilde{\mathbf{k}}$ forming an angle 42° with the X axis in the XZ plane. The pump polarization is extraordinary (in the XZ plane), and the two-photon polarization is ordinary (along the Y axis). The longitudinal mismatch is calculated using the Selmeier formulas [17]. The length L of the crystal is 0.1 cm. Such a small length is used to increase the width of the frequency-angular spectrum; otherwise, some parts of it would be not resolved in the figure. The integral of the spectrum S determines the effective field of zero-point vacuum fluctuations (26), which, in its turn, determines the intensity of collinear frequency-degenerate three-photon PDC from a pump with the wavelength 351 nm (Ar laser) and the same wave vector direction as the one of $\tilde{\mathbf{k}}$. The angle of scattering in Fig. 4 is in one-to-one correspondence with the transverse wave vector q from the equations given above. For simplicity, the factor $f^2(\Omega,\Omega_1)$ was neglected in this calculation. Numerical calculation shows that the central part of the frequency-angular spectrum in

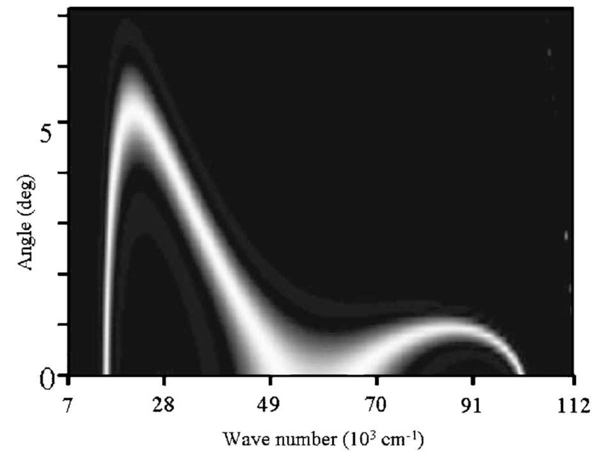

FIG. 4. Calculated frequency-angular spectrum of zero-point vacuum fluctuations "seeding" the collinear frequency-degenerate three-photon PDC in a lithium formate crystal of thickness 0.1 cm. The pump has the wavelength 351 nm and is directed in the XZ plane at an angle 42° to the X axis. Polarization of the three-photon light corresponds to the Y direction.

Fig. 4, which corresponds to near-collinear near-degenerate SPDC phase matching and for which the expansion (7) is valid, constitutes not the major part of the spectrum but approximately one-half.

For estimating the spectral width of the three-photon PDC, we have performed numerical integration of the spectra similar to the one shown in Fig. 4 at various "effective pump wavelengths" $\tilde{\omega}$. The result is shown in Fig. 5. To calculate each point in the figure, the observation wavelength λ_1 (and, hence, the observation frequency ω_1) was fixed, the corresponding wave vector $k_1(\omega_1)$ was calculated, and then the frequency and wave vector $\tilde{\omega}\equiv\omega_p-\omega_1$ and $\tilde{k}\equiv k_p-k_1$ were found. Using these parameters as the frequency and wave vector of the pump for two-photon PDC, the spectrum was calculated, similar to the one shown in Fig. 4. Then the spectrum was numerically integrated over the frequency and the angle, to obtain S, the integral spectrum of zero-point vacuum fluctuations "seeding" the three-photon PDC at

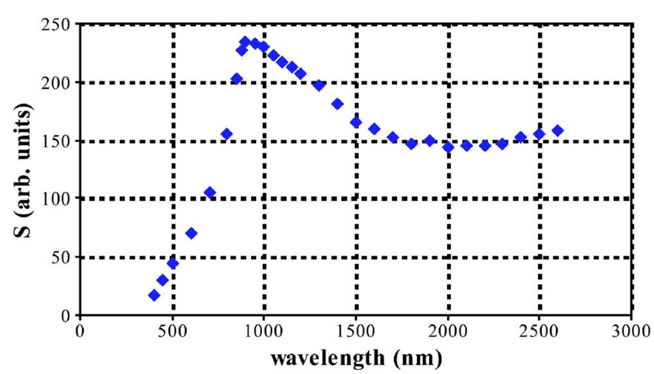

FIG. 5. (Color online) Integral of the spectrum of zero-point vacuum fluctuations "seeding" the three-photon PDC in lithium formate crystal calculated numerically as a function of the "observation wavelength" λ_1. The pump wavelength is 351 nm, and the pump wave vector is in the XZ plane at an angle 42° to the X axis.

wavelength λ_1. The dependence of S on λ_1 roughly [18] determines the intensity spectrum of three-photon PDC.

The spectrum shown in Fig. 4 corresponds to the degenerate case of three-photon PDC—i.e., to the situation where the wavelength of the registered photon λ_1 is exactly 3 times the wavelength of the pump (in our example, 1053 nm). From Fig. 5 one can see that the integral of the two-photon spectrum, and hence the intensity of three-photon light, is indeed close to being maximal in this case. However, the peak in the center, caused by the contribution from two-photon phase matching, is accompanied by a broad spectrum at large wavelengths. Although the intensity of three-photon PDC at large wavelengths will be reduced by the frequency-dependent factor $f^2(\Omega,\Omega_1)$, it is still clear that the spectrum of three-photon PDC in the collinear direction occupies a broad range from the pump wavelength to the far infrared.

It might seem that the results of the present section completely ruin the validity of the near-collinear near-degenerate approximation used in the previous sections. However, it is not so. First of all, the "near-collinear approximation" is still valid: calculation of the correlation functions was performed for the collinear case, the transverse wave vectors of the three photons being "filtered" by external apertures. Also, the angles of the two-photon PDC spectrum do not exceed 10°, as one can see from Fig. 4. As to the near-degenerate approximation, it is indeed not valid in general. However, the approach of Sec. IV can still be used in the case of zero or negative GVD at the wavelength λ_0, when the spectrum of SPDC is finite [19]. A similar approach can be also used if the three-photon light is partly filtered.

In addition, let us stress that the general approach used in the present section does not provide a "universal" analytical expression for the correlation functions or intensity distribution; it only enables one to perform numerical calculations for specific cases.

Finally, let us estimate the number of photons per mode that can be generated via three-photon parametric down-conversion. Taking the value of cubic susceptibility $\chi^{(3)}=10^{-13}$ esu, which is typical for nonlinear crystals like lithium niobate or KDP, the pump intensity $I_0=1$ kW/cm^2, the wavelength of the down-converted photons 1 μm, the crystal length $L=10$ cm, and the width of the frequency–wave-vector spectrum of the two-photon phase matching $\Delta^2\mathbf{q}\Delta\Omega=10^{18}$ s^{-1} cm^{-2}, from Eq. (24) we obtain for the three-photon down-conversion $N\sim 10^{-17}$. This figure is rather small; for instance, the typical number of photons per mode for two-photon SPDC is ten orders of magnitude higher. To pass from the number of photons per mode, N, to the counting rate W of a detector, one can use the relation [1]

$$W = NS^{\mathrm{vac}}_{\lambda\Theta}\eta\Delta\lambda\Delta\Theta\Delta A, \tag{28}$$

where $S^{\mathrm{vac}}_{\lambda\Theta}=0.5955$ W/(Å cm^2 sr) is the vacuum brightness [1], η is the detector quantum efficiency, and $\Delta\lambda$, $\Delta\Theta$, and A are, respectively, the wavelength range, solid angle, and the area selected by the detector. Taking $\eta=0.5$, $\Delta\lambda=40$ nm, $\Delta\Theta=10^{-4}$ sr, and $A=10^{-2}$ cm^2, we obtain the counting rate W on the order of 10^{-2} s^{-1}. A reliably detectable counting rate should be at least two orders of magnitude higher. However, some hope is given by the possibility to use media with resonance values of $\chi^{(3)}$, optical fibers with large length, or spatially inhomogeneous media with specially increased width of two-photon phase matching.

Beside the low efficiency of three-photon PDC, another problem connected with its experimental observation is the existence of other nonlinear processes resulting in the emission of photons in the same spectral range. Among them, the strongest are two-photon SPDC and four-wave mixing. However, the first one can be eliminated by choosing a medium without $\chi^{(2)}$ and the second one has a different dependence on the pump intensity (quadratic rather than linear).

VI. CONCLUSION

The main conclusion of the present paper is that the three-photon PDC of pump photons with frequency ω_p and wave vector $\mathbf{k}_p$ into a single frequency–wave-vector mode ω, $\mathbf{k}$ is "seeded" by the whole spectrum of zero-point vacuum fluctuations corresponding to all possible two-photon processes with pump frequency $\widetilde{\omega}\equiv\omega_p-\omega$ and wave vector $\widetilde{\mathbf{k}}\equiv\mathbf{k}_p-\mathbf{k}$. Accordingly, the intensity of three-photon PDC is determined by the integral spectrum allowed by the phase matching of two-photon PDC (SPDC). (This does not mean that the medium should have nonzero quadratic nonlinearity, a necessary condition for SPDC.) It follows that the efficiency of three-photon PDC can be increased by choosing a medium with a broad spectrum of two-photon PDC.

If the $\chi^{(3)}$ tensor of the medium has several nonzero elements corresponding to the chosen polarization of one of the output photons, one should take into account all spectra of two-photon processes corresponding to the possible polarizations of the other photons.

The frequency width of the three-photon PDC at fixed angle of scattering is much larger than the SPDC width. It does not depend on the thickness of the nonlinear medium and covers all the visible and near-infrared ranges. This gives additional hopes for observing three-photon parametric down-conversion and shows that the best way to observe this process is to use broadband detection.

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