Three-photon generation by means of third-order spontaneous parametric down-conversion in bulk crystals

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Abstract
We investigate the third-order spontaneous parametric down-conversion process in a nonlinear media with inversion centers. Specifically, we analyze in details the three-photon differential count rate in unit frequency and angular regions, total count rate and measurement time for rutile and calcite crystals which have comparatively large cubic susceptibilities. Special attention is given to consideration of limited frequency and angular detection ranges in order to calculate experimentally available detection rate values.

Keywords: nonlinear crystals, central symmetry, cubic susceptibility

1. Introduction

Generation of photon-number (Fock) states of light is one of the main tasks in quantum optics. They are interesting not only from fundamental, but also from practical points of view because of their necessity for solving problems of quantum communications and linear optical quantum computations. While problems of single-photon and biphoton-state generation are well studied, the direct and non post-selective generation of higher-order Fock states is still an attractive challenge.

In this work we consider the problem of three-photon state generation. Non-classical properties of such states enable heralded emission of photon pairs [1–3] as well as preparation of three-body entangled states (for example, Greenberger–Horne–Zeilinger (GHZ) states [4, 5]).

There are several proposed solutions for the problem of three-photon generation such as cascaded or postselective second-order nonlinear processes [6–12] and formation of approximate photon triplets by SPDC photon pairs together with an attenuated coherent state [13]. All these approaches give relatively low photon generation rates (up to 45 min⁻¹ [8]) and have a big contribution of low-photon-number impurities.

On the other hand, the most natural way to generate three-photon states is a third-order spontaneous parametric down-conversion (TOSPC). Unlike the other techniques, it enables to generate a three-particle entanglement in continuous degrees of freedom, such as energy and momentum. This problem was previously studied theoretically, but to the best of our knowledge no experimental results were reported for direct spontaneous generation of triplets based on $\chi^{(3)}$. Only stimulated third-order parametric down-conversion was demonstrated by seeding triplet modes [14, 15].

There are two approaches for TOSPC generation: in bulk crystals [6, 16–18] or in optical fibers [19–24]. The bulk crystals allow satisfying simply the phase-matching condition using different polarization modes, but a spatial multimode structure of three-photon light and a limited crystal length complicate a high photon conversion probability and an effective detection. One can increase the interaction length and decrease the spatial mode number by using optical fibers. In this case phase-matching condition can be realized while the pump and three-photon light propagate in different spatial modes or by using the quasi-phase-matching. But a small mode overlap ($\sim 10^{-3}$ [24]) and a high absorption coefficient...
for the pump (in a visible and especially in UV case) also limit the generation rate.

Our work presents a theoretical description of the third-order parametric down-conversion in crystals. Special attention will be paid to crystals with inversion centers, having zero $\chi^{(2)}$ and hence prohibiting all three-wave processes. That is of great importance especially in the case of triple generation with seeding beams because three-wave processes are much more intense and may suppress generation of triplets as well as their detection.

The paper is organized as follows. In section 2 we evaluate the three-photon count rate in unit ranges of frequency and transverse wave vector and the integral count rate over all detectable frequencies and transverse wave vectors of scattered photons in a collinear degenerate regime of generation for the type-I and type-II phase-matching. In section 3 we get estimates of the minimal measurement time sufficient for distinguishing signal triple coincidences from noise ones. Then in section 4 our estimates are specified for two nonlinear crystals with inversion centers: calcite and rutile. And finally in section 5 we discuss obtained results.

2. Calculation of photon count rate

As the process of third-order SPDC (TOSPDC) is similar to two-photon SPDC, in this section we follow the approach developed by Klyshko for biphotons in [25], though somewhat extended for the case of triplets.

Let a pump photon in the mode $k_p, \omega_p$ be decaying for three photons in modes $k_1, \omega_1, k_2, \omega_2$ and $k_3, \omega_3$ and let a pump be a monochromatic plane-wave propagating along the z-axis. Under these assumptions the photon energy and transverse momentum are conserved and the non-conservation of the longitudinal momentum determines the phase mismatch $\Delta k_z$:  

$$\omega_1 + \omega_2 + \omega_3 - \omega_p = 0, \tag{1}$$

$$\vec{q}_1 + \vec{q}_2 + \vec{q}_3 = 0, \tag{2}$$

$$k_1 + k_2 + k_3 - k_p = \Delta k_z, \tag{3}$$

where $\vec{q}_i$ denote perpendicular components of $\vec{k}_i$.

In the second order of the perturbation theory TOSPDC is described by the Hamiltonian

$$H = \frac{1}{2} \int_V d^3p \sum_{k_1, k_2, k_3} \chi^{(3)} E_{k_1} E_{k_2} E_{k_3} \Delta{k}_i a_{k_1}^\dagger a_{k_2}^\dagger a_{k_3} E_{p}$$

$$\times \exp[i(k_p - \vec{k}_1 - \vec{k}_2 - \vec{k}_3)p - i(\omega_p - \omega_1 - \omega_2 - \omega_3)t]$$

$$+ \text{H.c.}, \tag{4}$$

where $E_p$ is the amplitude of the pump considered as a classical monochromatic plane wave, $V$ is the interaction volume,  

$$c \approx \sqrt{\frac{2\pi \hbar \omega}{v}}, \tag{5}$$

$v$ is the quantization volume and $a_{k_i}^\dagger$ are the photon creation operators for modes $k_i$ ($i = 1, 2, 3, p$).

The rate of transitions per unit spectral and transverse-wave-vector ranges of each photon $d\omega_i$ and $d\vec{q}_i$ is determined by the Fermi golden rule

$$R_{\omega_1 \omega_2 \omega_3} = \Gamma I^2 W_p \sin^2 \left( \frac{\Delta k_z}{2} \right) \times \delta^{(2)}(\vec{q}) \delta(\Omega), \tag{6}$$

where

$$\delta^{(2)}(\vec{q}) = \delta(q_{1x} + q_{2x} + q_{3x}) \delta(q_{1y} + q_{2y} + q_{3y}),$$

$$\Omega_i = \omega_i - \omega_p/3, \quad \delta(\Omega) = \delta(\Omega_1 + \Omega_2 + \Omega_3), \tag{7}$$

$$\Gamma = h \chi^{(3)}(\omega_1 \omega_2 \omega_3) / (c^2 (2\pi)^2 n n n),$$

$n_i$ are the refractive indices, $W_p$ is the pump power and $c$ is the speed of light.

To calculate the total three-photon generation rate, we have to integrate the differential rate of equation (6) over spectral and angular regions, restricted by features of the detection scheme:

$$R_I \approx IT^2 W_p, \tag{8}$$

where

$$I \equiv \int \sin^2 \left( \frac{\Delta k_z}{2} \right) \delta^{(2)}(\vec{q}) \delta(\Omega)$$

$$\times d\omega_1 \ d\omega_2 \ d\omega_3 \ \ d\vec{q}_1 \ d\vec{q}_2 \ d\vec{q}_3. \tag{9}$$

The above-mentioned restrictions of the integration ranges will be paid to crystals with inversion centers, having zero order parametric down-conversion in crystals. Special attention was paid to crystals with inversion centers: calcite and rutile. And finally in section 5 we discuss obtained results.

In the biphoton case one can calculate the total count rate, integrating along the phase-matching curve, defined by the equation $\Delta k_z(q_i, q_i = -q_i, \Omega_1, \Omega_2 = -\Omega_1) = 0$. This case is comparatively simple as there are only two independent integration variables. Below we will derive the expressions for the curves analogous to the biphoton phase-matching curve in the case of triplets for the type-I and type-II TOSPDC collinear degenerate regime of generation.

In order to do this, let us expand $\Delta k_z$ in powers of $\Omega_i, q_i$ and $q_0$ up to the second order:

$$k_z = \sqrt{k_1^2 - q_1^2}, \quad k_i = n(\omega_i) \frac{\omega_i}{c} \quad i = 1, 2, 3, \tag{10}$$

$$\Delta k_z = k_{z1} + k_{z2} + k_{z3} - k_p = [k_1 + k_2 + k_3]_0 - k_p$$

$$+ \sum_{i=1,2,3} \left( \frac{\partial k_i}{\partial q_i} \right)_0 q_i + \left( \frac{\partial k_i}{\partial \Omega_i} \right)_0 \Omega_i$$

$$+ \frac{1}{2} \left( \frac{\partial^2 k_i}{\partial q_i^2} \right)_0 q_i^2 + \frac{1}{2} \left( \frac{\partial^2 k_i}{\partial \Omega_i^2} \right)_0 \Omega_i^2. \tag{11}$$
and equation (11) are not equal and the decomposition of $\Delta k_z$ has the following form (we assume that the photons 1 and 2 are ordinary and the photon 3 is extraordinary):

$$\Delta k_z = \beta_s (\Omega_1 + \Omega_2) + \beta_o \Omega_3 + \alpha q_{3x},$$

$$+ \frac{1}{2} \gamma_{1x} (q_{1x}^2 + q_{2y}^2) + \frac{1}{2} \gamma_{3x} q_{3x}^2,$$

$$+ \frac{1}{2} \gamma_{1y} (q_{1y}^2 + q_{2x}^2) + \frac{1}{2} \gamma_{3y} q_{3y}^2,$$

where

$$\beta_s = \frac{\partial k_{12}}{\partial \Omega_1}, \quad \beta_o = \frac{\partial k_{12}}{\partial \Omega_3}, \quad \alpha = \frac{\partial k_{12}}{\partial q_{3x}},$$

$$\gamma_{1x} = \frac{\partial^2 k_{12}}{\partial q_{1x}^2}, \quad \gamma_{1y} = \frac{\partial^2 k_{12}}{\partial q_{1y}^2}, \quad \gamma_{3x} = \frac{\partial^2 k_{12}}{\partial q_{3x}^2},$$

$$\gamma_{3y} = \frac{\partial^2 k_{12}}{\partial q_{3y}^2}.$$
derivatives $\alpha_{\gamma_0}$, but in the $y$-direction $\partial k_y / \partial q_y = 0$ and, hence, the second-order derivatives $\gamma_{\gamma_0 \gamma_0}$ have to be retained.

By denoting $\gamma_{\gamma_0 \gamma_0} \approx \gamma_{\gamma_0}$, we get

$$\Delta k_y = (\beta_y - \beta_0) (\Omega_1 + \Omega_2) - \alpha_y (q_{1x} + q_{2x}) + \gamma (q_{1y}^2 + q_{2y}^2),$$

and the integral $I$ of equation (9) takes the form

$$I = \frac{2\pi}{|\gamma|} \int \sin\left(\frac{1}{2} \Omega_{+} + \frac{1}{2} q_{+} \Omega_{-} \right) d \Omega_{+} d \Omega_{-} d q_{+} d q_{-},$$

where

$$\Omega_{\pm} = \Omega_{1,2} \pm \Omega_{3}, \quad q_{\pm} = q_{1,2} \pm q_{3},$$

$$\alpha_{\pm} = \sqrt{2} \alpha_0, \quad \beta_{\pm} = \sqrt{2} (\beta_0 - \beta_0),$$

with the intervals of $q_{1,2}$ where $|\Delta k_y| < 2\pi$ estimated as $\Delta q_y^2 = 2\pi / |\gamma|$. Hence, the exact-phase-matching curve for type-II TOSPDC has the form $\beta_0 \Omega_1 - \alpha_0 q_y = 0$ (the green dotted line in figure 2(a)). Each point of the phase-matching area $\{\Omega_+, q_y\}$ corresponds to the ranges $\Delta q_-$ (figure 2(b)) and $\Delta \Omega_-$ (figure 2(c)), which can be found from phase-matching conditions:

$$\Delta q_- = 2 \left( q_{m} \sqrt{2} - |q_{+}| \right) = 2 \left( q_{m} \sqrt{2} - \left| \frac{\beta_{++}}{\alpha_{++}} \Omega_{+} \right| \right)$$

(shown as blue arrow on figure 2(b)),

$$\Delta \Omega_- = 2 \left( \Omega_{m} \sqrt{2} - |\Omega_{+}| \right)$$

(shown as red arrow on figure 2(c)). Similarly to the type-I case we approximate the integral over $q_+$ by the product of the integrand at the exact-phase-matching curve with the width of the phase-matching area $\Delta q_+ = 4\pi / \alpha_0$ and get

$$I \approx \frac{2\pi}{|\gamma|} \int \sqrt{2} \Omega_{\max} d \Omega_{+} \Delta \Omega_{-} \Delta q_+ \Delta q_-$$

$$= \frac{32\pi^2}{|\alpha_0 \gamma|^2} \int \sqrt{2} \Omega_{\max} d \Omega_{+} \cdot \Omega_{m} \sqrt{2} - |\Omega_{+}| d \Omega_{+} \cdot \left( \Omega_{m} \sqrt{2} - |\Omega_{+}| \right)$$

$$\times \left( q_{m} \sqrt{2} - \left| \frac{\beta_{++}}{\alpha_{++}} \Omega_{+} \right| \right),$$

(18)
where $\Omega_{\text{max}}, \Omega_{m}, \text{and } q_m$ are determined by detection ranges in frequency and angle shown in figure 2:

$$q_m = \frac{k^2 \Delta \theta_{\text{max}}}{\lambda_{\text{min}}}, \quad \Omega_{\text{max}} \equiv \min[\Omega_m, \Omega_q], \quad \text{with}$$

$$\Omega_m \equiv \min\left[\frac{2\pi c}{\lambda_{\min}}, \frac{\omega_p}{3}, \frac{\omega_q}{3}\right], \quad \Omega_q \equiv \frac{\alpha_+}{\beta_+}.$$

3. Evaluation of the measurement time

The measurement time $T_3$ can be defined as the time sufficient for extracting the signal triple coincidence count rate $R_3^{(3)}$ from noise $R_3^{(3)}$. Mathematically this means that the total number of triple coincidence counts (found as the difference of two measured rates, of the sum of signal and noise counts and, separately, of only noise counts) $N_s^{(3)} = R_3^{(3)} T_3$ is at least $t_{C,\infty}$ times bigger than its standard deviation $\sigma_s^{(3)}$, where $t_{C,\infty}$ is the Student’s $t$-factor for a confidence level C. Taking into account the Poisson distribution of photo counts one can calculate the dispersions:

$$\sigma_s^{(3)} = \sqrt{R_3^{(3)} T_3},$$

$$\sigma_{R_s}^{(3)} = \sqrt{(R_3^{(3)} + R_3^{(3)}) T_3},$$

$$\sigma_r^{(3)} = \sqrt{(\sigma_s^{(3)})^2 + (\sigma_{R_s}^{(3)})^2} = \sqrt{(2R_3^{(3)} + R_3^{(3)}) T_3}.$$

So, we obtain the equation for $T_3$:

$$N_s^{(3)} = T_3 R_3^{(3)} = t_{C,\infty} \sigma_s^{(3)} = t_{C,\infty}\sqrt{(2R_3^{(3)} + R_3^{(3)}) T_3}.$$

With given quantum efficiency $\eta$ and the noise count rate $R_3^{(3)}$ of each detector, we assume that all detectors have approximately the same characteristics, the temporal resolution of electronics (typically limited by a detector jitter) $\delta \tau$, and $R_T$ evaluated in (8), we get

$$R_3^{(3)} = (R_3^{(1)})^2 \delta \tau^2,$$

$$R_3^{(3)} = \xi R_T \eta^3,$$

where the parameter $\xi$ characterizes features of non-polarized beam splitters to be used in a possible experimental setup for dividing TOSPDС signal into three channels. Hence,

$$T_3 = t_{C,\infty} \frac{2(R_3^{(3)})^2 \delta \tau + \xi R_T \eta^3}{(\xi R_T \eta)^2}.$$

Similar expressions can be derived for the minimal time $T_2$, $T_1$ (or $T_2$) required for distinguishing two-photon signal coincidence and single-photon counts from the noise:

$$T_2 = t_{C,\infty} \frac{2(R_3^{(3)})^2 \delta \tau + \xi R_T \eta^2}{(\xi R_T \eta^2)^2},$$

$$T_1 = t_{C,\infty} \frac{2(R_3^{(3)})^2 \delta \tau + R_T \eta}{(R_T \eta)^2}.$$

*We consider an ideal case when the number of the noise counts equals to the number of intrinsic detector’s dark counts and the light noise is neglected.

4. Example: calcite and rutile crystals

Let us make estimates for two specific crystals, calcite and rutile, having comparatively large cubic susceptibilities. For these two crystals and for four pump wavelengths, 266, 325, 405 and 532 nm, the results of calculations are presented in table 1. By using data about crystal refractive indices of [26, 27], we found values of angles between the optical axes of crystals and the pump propagation direction providing collinear emission of TOSPDС photons. For these orientations of crystals, with the use of matrix elements determining $\chi^{(3)}$ [28–31], and with the dependence of $\chi^{(3)}$ on the angle between the pump propagation direction and the crystal optical axis [32] taken into account, we calculated values of the effective cubic susceptibility $\chi_{\text{eff}}^{(3)}$ for both crystals and for all collinear TOSPDС regimes indicated in table 1. Together with $\chi_{\text{eff}}^{(3)}$ we present in table 1 the total count rates (8), calculated from equation (15) for rutile and from equation (18) for calcite.

Note that though rutile is a positive crystal and in the type-I phase-matching all three TOSPDС photons are not ordinary, it can be shown that even in this case the calculation based on (15) results in accuracy of $\Delta k / l$, about 10 percents.

The phase-matching conditions in calcite at the mentioned pump wavelengths are satisfied for each type of phase-matching (e→ooo, e→ooo and e←ooo), but for all types except e→ooo values of $\chi_{\text{eff}}^{(3)}$ are very small and, therefore, these cases are not included into table 1.

We paid special attention to calculations of the optimal crystal length $l$ and angular detection range $\theta_{\text{max}}$.

Note, first, that the total count rate is proportional to $l$ in type-I (6) and (15) and independent of $l$ in type-II (6) and (18) phase-matching cases. The last statement seems to be counterintuitive, but takes place due to compensation of two opposite processes. On the one hand, the increase of crystal length leads to increasing the interaction volume and so provides with higher differential generation rate. But on the other hand, for a longer crystal we get stronger phase matching conditions, smaller phase matching area and therefore, smaller number of vacuum (fluctuation) modes, stimulating three-photon generation. For biphoton generation there is a similar situation, but the second factor contributes not so significant, due to smaller fraction of vacuum modes, participating in the process.

The exact compensation takes place while in the phase-matching inequality (see (17) and further)

$$|\Delta_k| = \left|\frac{ax + \frac{1}{2} y^2}{l}\right| < \frac{2\pi}{l},$$

the quadratic term can be omitted. For calcite this is true for $l\gg l_{\text{max}} = 4\pi y/ax^2 \sim 0.05$ mm. But the crystal length can limit the detection angular range. For multi-mode detection
Table 1. Calculated values of effective third-order nonlinear susceptibility $\chi^{(3)}$, triplets generation rates $R_T$, measurement times necessary for registration of triple $T_1$ and double $T_2$ coincidences for different pump wavelengths $\lambda_p$ and power $W_p$, type of nonlinear media, its length $l$ and type of phase-matching, for different detectors and for the cases with presence (+) and absence (−) of the cavity.

| $\lambda_p$ (nm) | Medium | $\chi^{(3)}_e$ $(10^{-15}$ esu$)$ | $W_p$ (W) | Detector | l (mm) | Cavity | $R_T$ (Hz) | $T_1$ (days) | $T_2$ (days) |
|------------------|--------|-----------------------------------|-----------|----------|--------|--------|------------|------------|------------|
| 266              | Calcite| 0.32                              | 10        | Si APD   | 0.1    | −      | 4.0 $\cdot$ 10^{-5} | 94         | 15         |
|                  |        | (e → ooe)                         |           |          |        |        | 4.0 $\cdot$ 10^{-2} | 9.4 $\cdot$ 10^{-2} | 1.4 $\cdot$ 10^{-2} |
| 325              | Calcite| 0.59                              | 0.05      | Si APD   | 0.1    | +      | 1.1 $\cdot$ 10^{-5} | 5 200      | 750        |
| (e → ooe)        |        | Super Cond.                       |           |          |        |        | 3.4 $\cdot$ 10^{-6} | 18 000     | 1 000      |
| 405              | Calcite| 0.76                              | 0.5       | PMD      | 0.1    | +      | 1.8 $\cdot$ 10^{-4} | 8.1 $\cdot$ 10^{10} | 2.0 $\cdot$ 10^{11} |
| (e → ooe)        |        | Super Cond.                       |           |          |        |        | 9.5 $\cdot$ 10^{-6} | 6 200      | 370        |
| 532              | Calcite| 0.88                              | 10        | PMD      | 0.1    | +      | 1.8 $\cdot$ 10^{-4} | 8.2 $\cdot$ 10^{10} | 2.0 $\cdot$ 10^{11} |
| (e → ooe)        |        | Super Cond.                       |           |          |        |        | 5.1 $\cdot$ 10^{-6} | 12 000     | 690        |
| 532              | Rutile | 71.6                              | 10        | PMD      | 100    | −      | 1.5 $\cdot$ 10^{-2} | 1.1 $\cdot$ 10^{7}  | 2.8 $\cdot$ 10^{7}  |
| (o → eee)        |        | Super Cond.                       |           |          | 0.77   |        | 8.9 $\cdot$ 10^{-7} | 6.6 $\cdot$ 10^{4}  | 3.9 $\cdot$ 10^{3}  |

Note: Easy accessible in an experiment values marked as green, hardly accessible as yellow and unaccessible as red.

Table 2. Characteristics of single-photon detectors to be used for three-photon registration.

| Type                  | $\lambda_{min}$−$\lambda_{max}$ (nm) | Number of spatial modes | Quantum efficiency $\eta$ | Dark count rate $R_0^{(1)}$ (Hz) | Jitter $\delta \tau$ (ps) |
|-----------------------|-------------------------------------|-------------------------|---------------------------|-------------------------------|-------------------------|
| Si APD$^a$            | 400–1040                            | Multi                   | 0.1–0.7                   | 100                           | 350                     |
| InGaAs APD$^b$        | 1000–1650                           | Single                  | 0.1                       | 3000                          | 200                     |
| Super conductive$^c$  | 600–1700                            | Single                  | 0.2                       | 1                             | 50                      |
| PMD$^d$               | 950–1700                            | Multi                   | 0.01                      | 50 000                        | 70                      |

$^a$ Excilita SPCM-AQRH-16.

$^b$ IDQuantique ID210.

$^c$ Scient SSPD.

$^d$ Hamamatsu R3809U-69.

(see table 2) we assume $\theta_{max} = \pi/2^5$ (we suppose, that all the SPDC radiation can be focused on the detector area). But for single-mode detection the angular range is determined by a gaussian mode divergence $\theta = \lambda/\pi w$, where $w$ is the waist of the pump, which should be less than the spatial walk-off $pl$ (for the considered crystals $\rho \sim 0.1$). So, we obtain $\theta_{max} \propto l/\ell$. It means that in the case of type-II phase-matching we need to use as thin crystal as possible (we set $l = 2 \ell_{min} = 0.1$ mm). In the case of type-I phase-matching the crystal length $l$ has to be decreased until the integration limits in equation (15) become defined by the angular range. This means that the optimal crystal length is

$$l = \frac{k_p \lambda}{3 \pi \rho} \sqrt[\nu]{\frac{\alpha}{\beta}} \min \left[ 2 \pi c \frac{\omega_p + \omega_p}{3}, \frac{\omega_p - 2 \pi c}{l_{max}} \right].$$

Finally, for multi-mode detection and the type-I phase-matching the crystal has to be taken as long as possible (we set 100 mm).

5. Results and discussion

In calculations we took parameters of the most widely used single-photon detectors (see table 2) and the most suitable cw lasers: DPSS lasers at 266 and 532 nm, diode blue-ray laser at 405 nm and HeCd gas laser at 325 nm. Typical powers are given in table 1. It was shown [22] that the use of pulsed pump lasers gives no advantages for TOSPDG generation.

Also we consider a possibility of increasing the pump power inside the crystal by means of mirror deposition at the rear and front faces of the crystal, which turns the latter into a cavity. Accordingly, maximal growth of the pump intensity in a crystal (cavity) is characterized by the factor $\varepsilon = 1000^6$. Unfortunately, this optimization cannot be used in rutile crystals because of their comparatively high adsorption.

In addition to the total triplet generation rates $R_T$, we have calculated and presented in table 1 the time, required for two- and three-photon coincidences, (21) and (22) at $t_0_{99.9\%} = 3$.

One can see, that one of the main problems of TOSPDG detection is related to low efficiency and high noise of IR detectors. It is really difficult to extract a signal from noise even for triple coincidence measurements. So we found only one combination of parameters when detection of TOSPDG photons looks possible. This is the case of a calcite crystal with a mirror coverage deposited at rear and front faces, the crystal length $l = 0.1$ mm, and the pump parameters $\lambda_p = 266$ nm and $W_p = 10$ W. In this case we found $T_2 = 20$ and $T_3 = 135$ min.

6 According to [33], the reflection coefficient of a lossless mirror is $R_M \approx 0.999$, which gives $\varepsilon \approx (1 - R_M)^{-1} \approx 1000$. Losses in mirrors can decrease $\varepsilon$ making it not higher than 10 for existing AR coatings.

5 Of course, so big angles are outside of the framework of our model but we suppose that, still, it remains reasonable for rough estimates.
One more advantage of UV pump is the increase of the differential generation rate, because it is proportional to $\omega_{\Delta}^{3/2}$, or $\chi_{\text{eff}}^{(3)}$ (6).

Another well-known problem is a low value of $\chi_{\text{eff}}^{(3)}$. For rutile crystal $\chi_{\text{eff}}^{(3)}$ is about two orders higher if the crystal optical axis is taken parallel to the pump propagation direction. But for the phase-matching conditions to be satisfied one has to use a crystal with periodical poling (quasi-phase-matching). This is an evident way for making the type-I and type-II phase-matching conditions satisfied and TOSPDC photons detectable in visible range of wavelengths. Another possibility of increasing $\chi_{\text{eff}}^{(3)}$ is an addition of special impurities to crystals which would provide resonance enhancement of the third-order susceptibility.

One more problem is a really multi-mode structure of TOPDC generation in bulk crystals, which requires multi-mode detection of signals. This problem can be solved by producing a wave-guide inside the crystal medium, as proposed in [34]. In this way the length of interaction can be done arbitrary long.

6. Conclusion

Finally, we have performed a detailed analysis of three-photon generation in birefringent crystals with special attention paid to calcite and rutile crystals. The analysis includes the calculation of differential generation rate in unit frequency and transverse wave vectors range (6), total count rate (8) for type-I and type-II phase-matching and measurement time, required for distinguishing signal coincidences from noise ((22) and (21)).

The results show that the registration of TOSPDC in calcite is possible for the process with the pump at 266 nm with the presence of a cavity. All the other considered cases need too much time for three-photon registration.

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