Characterization of parametric down-conversion in periodically-poled KTP crystals with a picosecond pump

S. Cigdem Yorulmaz and Michiel J.A. de Dood
Huygens Laboratory, P.O. Box 9504, 2300 RA Leiden, The Netherlands

ABSTRACT

We experimentally characterize sources of frequency degenerate down-converted photons at 826.4 nm generated in 2 mm, 5 mm and 10 mm long periodically-poled KTP crystals. The crystals are pumped by 413.2 nm laser pulses with 2 ps duration. The dispersion $D = 1.3$ ps/mm puts a limit to the length over which phase matching is efficient for a 2 ps pulse and provides a lower limit for the angular width of SPDC in the far-field. We investigate the far-field distribution of SPDC produced by periodically-poled KTP crystals and compare this with the calculated intensity distribution and find good agreement with theory. We also discuss the performance of PPKTP in terms of nonlinearity and group velocity walk-off compared to other available materials.

Keywords: Spontaneous parametric down conversion, periodically-poled KTP crystal, group velocity walk-off

1. INTRODUCTION

Spontaneous parametric down-conversion (SPDC) is a second order nonlinear process that converts a high energy pump photon into two highly correlated signal and idler photons at lower energy. Because of the strong correlations between the photons, SPDC is a good candidate for creating entangled photons. Sources of polarization, frequency, time-bin and spatially entangled photons all have been demonstrated.

Here we consider spatially entangled photons with the same polarization, which can be described by the transverse photon momentum. Generating such spatially entangled single photon pairs has been demonstrated using continuous wave lasers as a pump. However, in order to create spatially entangled multi-photon states, the photons should be generated in a single time-bin and the ultra short coherence time of the photon pairs should be compensated by the use of short laser pulse. In this paper, we present experimental characterization of periodically-poled KTP crystals with different length pumped by a picosecond pulsed laser. We discuss the effects of group velocity walk-off length in PPKTP using the group velocity mismatch. To compare the characteristics of pulsed SPDC source, we compare our results to the calculated intensity distribution of a continuous wave pumped SPDC source based on PPKTP. We also discuss the efficiency for pulsed SPDC of different crystal types such as PPLN, BBO, LBO and LiIO$_3$ compared to PPKTP crystal.

2. PHASE MATCHING IN PULSED DOWN-CONVERSION

In parametric down-conversion process, energy is conserved, so the frequencies of the photons are related via $\omega_p = \omega_s + \omega_i$; where $p$, $s$ and $i$ correspond to pump, signal and idler photons, respectively. For an efficient process, photons are expected to satisfy the phase matching condition $\overrightarrow{k}_p = \overrightarrow{k}_s + \overrightarrow{k}_i$ where $\overrightarrow{k}$ represents the wavevector of the photons. In this article, we focus on frequency-degenerate SPDC generated in periodically poled KTP crystals for which signal and idler photons have the same frequency ($\omega_p = 2 \omega_s = 2 \omega_i$). To simplify the notation, we use $k_{2\omega}$ and $k_\omega$ for the wavenumber of the pump and SPDC photons. Phase matching for continuous-wave pumped SPDC is well known and has been extensively described for periodically-poled KTP. For a pump beam propagating in the $x$ direction of the crystal, the transverse wavevector mismatch in $y$-$z$ plane is approximately zero i.e. ($q_x \approx -q_z$), while the longitudinal wavevector mismatch is not zero due to the temperature-dependence of the refractive index of the PPKTP crystal. The intensity distribution of continuous-wave SPDC can be written as

$$I(q, T) \propto \text{sinc}^2 \left( \frac{L}{k_{2\omega} q_x^2 + \varphi(T)} \right)$$

(1)
where $q_\omega$ is the transverse momentum of down-converted photon, $L$ is the crystal length and $\varphi(T)$ is a temperature-dependent longitudinal phase mismatch. Collinear SPDC corresponds to $\varphi(T) = 0$ and occurs only for a specific crystal temperature.

We consider here frequency-degenerate down-conversion generated by short, picosecond, laser pulses in the same PPKTP crystal. Since a pulsed pump has a nonzero spectral width, the phase matching condition should be modified to contain the effect of this spectral width of the pump. We use a first order Taylor expansion of the wavevector mismatch for frequency-degenerate SPDC to include the effect of dispersion on the phase matching condition

$$\Delta k_x(\Omega) \approx (k_x(2\omega) - 2k_x(\omega)) + D\Omega$$

(2)

Here $D$ is the dispersion that contains the group velocity mismatch between pump and down-converted photons and is defined as $D = 1/v_g(2\omega) - 1/v_g(\omega) = (n_g(2\omega) - n_g(\omega))/c$ with $v_g$ and $n_g$ the group velocity and group index. The frequency $\Omega$ is the detuning of the pump frequency relative to the center frequency of the pump. Note that to arrive at Eq.2, we have used the fact that pump and down-converted photons all have the same polarization, and that we neglect the relatively small phase-mismatch due to differences in the frequency of the SPDC photons. This second simplification corresponds to a typical experimental situation where the SPDC light is filtered by a narrow bandpass filter with a bandwidth much smaller than the natural bandwidth of the source. For comparison, the natural phase matching bandwidth for a 2 mm PPKTP crystal is around 40 nm, while the bandpass filter has a FWHM width of 1.5 nm. For PPKTP the difference in group index $n_g(2\omega) - n_g(\omega) = 0.4564$ at a crystal temperature of 300K and pump wavelength of 413.2 nm gives $D = 1.3 \text{ ps/mm}$.

By assuming a Gaussian envelope function to represent the frequency content of the pulsed pump, the intensity distribution for pulsed down-converted photons generated in PPKTP can be written as

$$I(q, T) \propto \frac{1}{\sigma \sqrt{2\pi}} \int d\Omega \exp\left(-\frac{1}{2}(\Omega/\sigma)^2\right) \text{sinc}^2\left(\frac{L}{k_\omega^2} q_\omega^2 + \varphi(T) + \frac{1}{2}D\Omega L\right)$$

(3)

where $\sigma$ is a measure of the spectral width of the pulse. For efficient conversion of all spectral components in the pulse, the condition of $DL < \tau_p$ has to be satisfied, where $\tau_p$ is the duration of the pulse. The group velocity walk-off length $L_w = \tau_p/D$ is a typical length over which the SPDC process is efficient. For crystals longer than this walk-off length, the pulse duration of the SPDC light is significantly broadened compared to the pump pulse.

In order to visualize the role of group velocity walk-off in pulsed down-conversion processes, we calculate the angular width of collinear SPDC ring as a function of crystal length for both continuous wave (Eq.1) and pulsed laser pump (Eq.3). Figure 1 compares the calculated angular width (FWHM) of collinear SPDC for continuous wave (red line) and a pulsed pump (black line) with a pulse duration $\tau = 2$ ps ($\sigma = 2.74 \times 10^{-12}$ s$^{-1}$), as a function of crystal length on a log-log scale. For continuous-wave pumping, the angular width decreases inversely proportional to $\sqrt{L}$. For pulsed laser pumping, the width saturates when the crystal is longer than walk-off length $L_w = 1.3$ mm. For short crystals ($L < 0.15$ mm), the effect of dispersion can be ignored and the phase matching conditions for continuous wave and pulsed SPDC become identical. For comparison, the inset of Figure 1 shows the calculated angular intensity distributions of SPDC photons created in 1.3 mm long PPKTP crystal for collinear-phase matching using a pulsed (black line) and continuous-wave (red line) pump.

3. EXPERIMENTAL CHARACTERIZATION OF PULSED SPDC SOURCE

In our experiments, we used periodically-poled KTP crystals with different lengthS pumped by a weakly focused ($\omega_p = 100\mu m$), vertically polarized pulsed laser propagating along the x-axis of the crystals. The length of the crystals are 2.0 mm, 5.0 mm and 10.0 mm in the x-direction and all crystals have the same width of 2 mm and height of 1 mm. The poling period of the PPKTP crystal $\Lambda_0 = 3.675 \mu m$ is designed to enable collinear phase matching at a crystal temperature of 50°C to generate vertically polarized SPDC light at the frequency degenerate wavelength of 826.4 nm.

Figure 2 shows the experimental setup to obtain far-field images of SPDC light. 2 ps pulses from a frequency-doubled Ti:Sapphire laser at a wavelength of 413.2 nm are focussed into a periodically-poled KTP crystal to create non-collinear, frequency degenerate down-converted photons at 826.4 nm. A GaP wafer and a bandpass
Figure 1. Calculation of angular width (FWHM) of collinear down-converted light as a function of crystal length for both continuous wave and pulsed SPDC process. A spectral width $\sigma = 2.74 \times 10^{12}$ s$^{-1}$ for 2 ps pulses is taken into account to calculate the width for pulsed SPDC. Dashed lines indicate the limits of angular width and group velocity walk-off length $L_w$ where the pulsed curve is saturated for the crystal length longer than 1.3 mm. The inset shows calculated intensity versus angle of collinear SPDC light generated by continuous wave and pulsed laser for a crystal length of $L = L_w = 1.3$ mm.

filter (BF) with a 1.5 nm FWHM bandwidth are used to filter the pump beam and spectrum of SPDC light. The two lenses with focal distance of 50 mm and 25 mm are used to create a far-field image of the down-converted light on the CCD camera. The temperature of the PPKTP crystal is controlled via a Peltier element with a PID controller, which keeps the temperature constant within 10 mK. Temperature-dependent far-field images of the SPDC source are recorded by a CCD camera for the temperature range of 10-50°C.

Figure 2. Schematic of the experimental setup to record far-field images of down-converted light produced by a PPKTP crystal. The PPKTP crystal is pumped with pulsed frequency-doubled Ti:Sapphire laser producing 2 ps pulses at 80 MHz repetition rate.

The temperature dependence of the refractive index of the PPKTP crystal determines the angular distribution of the generated SPDC light in the far-field through the temperature dependent phase-mismatch $\varphi(T)$. Figure 3 shows CCD images of the far-field intensity distribution of SPDC light at temperatures of 10°C, 30°C and 50°C generated in a 2 mm long PPKTP crystal. The pixel numbers were converted to far-field angles using the size of the pixels of the CCD camera and focal length of the second lens $f = 25$ mm that generates the far-field image. The down-converted photon emission is close to collinear phase matching ($\varphi \approx 0$) at the crystal temperature of
50°C, while at 10°C, the collected SPDC photons in the far-field form an open ring corresponding to negative phase mismatch ($\varphi < 0$). In comparison to continuous-wave pumped SPDC light, the intensity distribution of collinear SPDC has a shape that can be described by a Gaussian rather than the typical sinc shape of Eq.1 for continuous wave pump.

We recorded the intensity distribution of SPDC photons at 826.4 nm in the far-field from 2 mm, 5 mm and 10 mm long PPKTP crystals as a function of temperature between 10 and 50°C. In order to find the width (FWHM) and radius of the SPDC ring, each image is first converted to polar coordinates around its approximate center and then azimuthally averaged to obtain a radial distribution. The exact center is found by a numerical search that calculates the radial distribution as a function of center position to find the exact center that maximizes the peak intensity and minimizes the width. Figure 4 shows the radial distribution for crystal temperatures of 10°C (top, circles), 30°C (middle, squares) and 50°C (bottom, triangles). To determine the temperature dependent phase mismatch $\varphi(T)$, the radial distribution for each crystal and all temperatures are fitted to Eq.3. Typical examples of the fit are shown by the solid lines in Figure 4.

Figure 5 shows the temperature dependence of the phase mismatch $\varphi(T)$, which is related to the known temperature-dependent refractive index of PPKTP crystal,10,14 In order to describe the results in Figure 5 and compare them to literature, we introduce a Taylor expansion for the temperature dependent phase mismatch around the temperature $T_c$ where collinear phase matching is obtained.

$$\varphi(T) = -\frac{1}{2}k_p L[\alpha(T_c - T) - \beta(T_c - T)^2]$$ (4)

For non-collinear SPDC, the negative phase mismatch provides a measure of the opening angle of the SPDC ring. Figure 5 shows the phase mismatch values extracted from the fit for the 2 mm (square), 5 mm (circles) and 10 mm (triangle) long PPKTP crystals as a function of temperature. The solid lines in Figure 5 correspond to fits of the measured phase mismatch to Eq.4. The coefficients that we obtained from our measurements are $\alpha = (23.7 \pm 0.3) \times 10^{-6}$ °C$^{-1}$, $\beta = (6.6 \pm 0.8) \times 10^{-8}$ °C$^{-2}$ and $T_c = 49.2$°C for the 2 mm crystal. For 5 mm PPKTP, we find $\alpha = (24.1 \pm 0.8) \times 10^{-6}$ °C$^{-1}$, $\beta = (4.7 \pm 0.9) \times 10^{-8}$ °C$^{-2}$ and $T_c = 52.5$°C. For the 10 mm crystal, the coefficients are $\alpha = (23.3 \pm 0.3) \times 10^{-6}$ °C$^{-1}$, $\beta = (6.0 \pm 0.8) \times 10^{-8}$ °C$^{-2}$ and $T_c = 49.3$°C. Coefficients measured with continuous wave pump using 5 mm PPKTP crystal10 are $\alpha = (24.0 \pm 0.2) \times 10^{-6}$ °C$^{-1}$ and $\beta = (4.8 \pm 0.3) \times 10^{-8}$ °C$^{-2}$ using a reference temperature of 25°C. In literature, according to the explicit expression for the temperature dependent refractive index of PPKTP crystal,14 the coefficients are calculated to be $\alpha = 23.56 \times 10^{-6}$ °C$^{-1}$ and $\beta = 8.6 \times 10^{-8}$ °C$^{-2}$. Our values of $\alpha$ are consistent with this earlier work on PPKTP while $\beta$ is somewhat lower, consistent with previous work for continuous wave pumping.10 From data in Figure 5 it is clear that the 5 mm PPKTP crystal has a slightly different collinear phase matching temperature compared to the 2 mm and 10 mm long crystals, which we believe, is due to the small variations in the optical properties.
Figure 4. Radially integrated intensity of recorded far-field images from Fig(3) as a function of far-field angle for a crystal temperature of 10°C (circle), 30°C (square) and 50°C (triangle). Solid lines are fits of Eq.3 to the data (see text). Collinear phase matching is obtained close to the crystal temperature of 50°C.

Figure 6 shows the temperature dependence of the angular width (FWHM) of the SPDC ring in pulsed down-conversion for three different crystal lengths by plotting the angular width as a function of phase-mismatch. We obtained the angular width of SPDC rings by fitting the radial intensity distribution by Eq.3 using $\phi$ and $\sigma$ as fitting parameters. In Figure 6, results for the 2 mm, 5 mm and 10 mm PPKTP crystals are shown by triangle, circle and square symbols, respectively. The fact that the value of spectral width of the pulse $\sigma$ varies for different crystal length may indicate that the CCD images are not exact far-field images. For the different PPKTP crystals, we find a spectral width of the pulse $\sigma$

<table>
<thead>
<tr>
<th>Crystal</th>
<th>$n_g(2\omega) - n_g(\omega)$</th>
<th>$D$ (ps/mm)</th>
<th>$d_{\text{eff}}$ (pm/V)</th>
<th>$L_wd_{\text{eff}}^2$ (nm$^3$/V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PPKTP</td>
<td>0.456</td>
<td>1.5</td>
<td>9.8</td>
<td>125</td>
</tr>
<tr>
<td>PPLN</td>
<td>0.514</td>
<td>1.7</td>
<td>24.3</td>
<td>710</td>
</tr>
<tr>
<td>BBO</td>
<td>0.053</td>
<td>0.18</td>
<td>2.0</td>
<td>45</td>
</tr>
<tr>
<td>LBO</td>
<td>0.033</td>
<td>0.11</td>
<td>0.8</td>
<td>11</td>
</tr>
<tr>
<td>LiIO$_3$</td>
<td>0.154</td>
<td>0.51</td>
<td>3.4</td>
<td>45</td>
</tr>
</tbody>
</table>

Table 1. Group index mismatch $n_g(2\omega) - n_g(\omega)$, dispersion $D$ and effective nonlinearity $d_{\text{eff}}$ for different crystals.
For a crystal longer than the walk-off length $L_w = \tau/D$, the efficiency of the non-linear process of SPDC is lowered. Hence, to create a large number of photon pairs it is important to consider the combined effect of the nonlinearity and the effect of walk-off and raises a question which crystal should be used to create spatially entangled multi-photon state by SPDC. As can be seen from the table, the effective non-linearity of the crystals is strongly correlated with the dispersion $D$. This empirical fact is well-known as Miller’s rule.\textsuperscript{11,15} The total number of photon pairs created by SPDC at the degenerate wavelength and integrated over the entire angular range of the SPDC cone is proportional to the single pass gain of the crystal. Since the useful length for efficient SPDC is given by the walk-off length a good number to compare the pair production rate of different crystal is $L_w d_{\text{eff}}^2$. As can be seen, PPKTP and PPLN are expected to outperform sources based on BBO or LiIO\textsubscript{3}. It should be noted that the birefringent phase-matching required for BBO, LBO and LiIO\textsubscript{3} also introduces a spatial walk-off that depends on the waist of the pump beam. This spatial walk-off could shorten the length over which phase-matching is efficient and the number given in the table should be regarded as an upper limit for these crystals. For BBO the walk-off angle $\rho$ is as large as 66 mrad, limiting the useful length for a realistic 100 $\mu$m pump beam to 1.5 mm. It is interesting to note that the walk-off angle in LBO is only 16 mrad, making the performance of LBO comparable to that of BBO when spatial walk-off becomes important. Periodically poled crystals such as PPKTP and PPLN do not suffer from spatial walk-off since the polarization of the pump and downconverted photons are identical. This greatly simplifies the design of the source and interpretation of the data. Given the large non-linearity of PPLN compared to PPKTP and a rather similar value of $D$, PPLN crystals are clearly preferred. This is especially true for processes that involve higher photon numbers as the yield of double pairs and triple pairs scales non-linearly with the efficiency of the source.

In conclusion, we have characterized a pulsed source of frequency degenerate photon pairs created by SPDC in 2 mm, 5 mm and 10 mm long PPKTP crystals pumped by 2 ps laser pulses at 413.2 nm. We have compared our experimental results with calculations of intensity distributions of continuous-wave and pulsed SPDC. The effect of dispersion on the phase matching condition due to a pulsed pump is well described by a calculations. For crystal lengths longer than the group velocity walk-off length $L=1.3$ mm, the efficiency of the source drops and the angular width of the far-field distribution of the SPDC light saturates.
Figure 6. Angular width (FWHM) of SPDC rings obtained from fitting the radial intensity distribution by Eq.3 as a function of phase mismatch for 2 mm (square), 5 mm (circle) and 10 mm (triangle) PPKTP crystal. The corresponding crystal temperature range is 10 – 50°C for all crystals. The dashed lines correspond to the spectral width of the pulse predicted by Eq.3 using spectral widths $\sigma_{2\text{mm}} = 2.66 \times 10^{12}s^{-1}$, $\sigma_{5\text{mm}} = 2.6 \times 10^{12}s^{-1}$ and $\sigma_{10\text{mm}} = 2.42 \times 10^{12}s^{-1}$. Data is fitted by Eq.3 (dashed line).

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