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**Title:** Beyond photon pairs  
**Issue Date:** 2014-06-10
The role of spatial and temporal modes in pulsed parametric down-conversion

We explore spatial correlations created by stimulated pair emission in frequency degenerate parametric down-conversion from a periodically poled KTP crystal pumped by ∼2 ps duration laser pulses. The ratio of stimulated pairs over spontaneous pairs reaches as high as 0.8 in the experiment. This ratio is a direct measure of the total number of modes relevant to the down-conversion process. We identify a universal curve for this ratio that accounts for the effect of the focused pump, introducing a coherence diameter $r_0$ related to the diffraction limited size of the pump beam in the far-field. Measurements of the spatial correlations of the PDC light for longer crystals and tight focusing conditions show that the description given in terms of a universal curve is surprisingly robust and breaks down only for a laser beam focused to a waist smaller than 40 µm in a 2 mm long PPKTP crystal.

S. C. Yorulmaz, M. P. van Exter, and M. J. A. de Dood, *The role of spatial and temporal modes in pulsed parametric down-conversion*, Optics Express 22, 5913-5926 (2014).
4 The role of spatial and temporal modes in pulsed parametric down-conversion

4.1 Introduction

It is well-known that parametric down-conversion (PDC) produces highly correlated quantum states that consist of pairs of photons. The quantum correlations of the pairs can pertain to the temporal [1–3], polarization [4–7] and/or spatial [8–11] degrees of freedom. Although polarization entanglement provides the paradigmatic system it is somewhat limited because each photon lives in a two-dimensional Hilbert space. In recent years other types of photonic entanglement have become almost as popular because they can provide a dimensionality per photon that is much larger than two. This high-dimensional aspect enhances the information-carrying capacity of the quantum system. As a result, improved security in quantum key distribution [12, 13], quantum coin tossing [14], increased quantum channel capacity [15, 16] and robust quantum imaging [17] have all been proposed and realized.

In parallel, multi-photon states of light have been investigated as they are an interesting resource because they can be used to test quantum mechanics in ways beyond what is possible with two qubits [18, 19]. The generation and control of more than two photons is a minimal requirement for possible future linear-optics quantum-computation [20] and may be useful for quantum communication protocols that are extra secure or robust against photon loss [21].

The combination of higher-dimensional quantum states with more than two photons is difficult to realize and characterize experimentally and has therefore received limited attention. To date, most experimental efforts use photons that are distinguishable in the time-domain and rely on measuring and modifying small differences in arrival time between photons. Spatial entanglement of photons provides an attractive alternative for higher-dimensional entanglement because the quantum correlations can be modified and detected with high (spatial) resolution simply by using lenses and apertures. The dimensionality of a spatially entangled state created by PDC depends on the crystal length and the beam waist of the focused pump [22, 23]. Non-collinear PDC is attractive because the photons within a pair are emitted in distinct directions, which greatly facilitates experiments because the photons can be detected on separate single photon counting detectors. In this way, spatial correlations of two-photon states have been demonstrated in a
variety of experiments, including quantum imaging [8, 24, 25], two-photon speckle [26, 27] and non-local momentum-position correlations [10, 28].

Thus far, most experimental work on spatial entanglement has addressed spatial quantum correlations at the two-photon level using a continuous wave pump laser to create individual photon pairs. Recently, we have explored spatially entangled four-photon states in an experiment with short pump laser pulses [29] and distinguish between double pairs created by spontaneous emission and true four-photon states created by stimulated emission [29]. Analogous to the case of time-bin entanglement [30], we introduced a visibility parameter $\chi$ that quantifies the relative importance of the stimulated-emission process. This parameter is a measure of the number of modes involved in the down-conversion process and plays a central role as it determines the photon number statistics of the quantum state created.

Here we explore how the visibility of the four-photon state, and hence the number of modes involved, depends on the focusing conditions and crystal length. By narrow spectral and spatial filtering, we are able to increase the visibility to values as high as 80%. For a constant crystal length, we find a universal curve for the visibility of the four-photon state as a function of the size of the detection aperture that combines all data for different pump beam diameters.

### 4.2 Properties of four-photon states produced by parametric down-conversion

The regime of parametric down conversion where multiple photon pairs are produced simultaneously is typically achieved using a pulsed, ultraviolet laser as a pump source. Both photon pairs can be generated via the spontaneous parametric down-conversion process. For this independent process the probability to create four photons is predicted by Poisson statistics and the corresponding probability is given by $P_4 = P_2^2 / 2$, where $P_2$ is the probability to produce a single pair via spontaneous parametric down-conversion. The process of stimulated parametric downconversion enhances the probability $P_4$. In an experimental configuration where only a single spatial and temporal mode is available, the process of spontaneous and stimulated emission are indistinguishable and the probability of spontaneous emission of a second pair exactly equals the probability of stimulated pair emission so that
$P_4 = P_2^2$. A visibility parameter $\chi$, ranging between 0 and 1, can be introduced \cite{30} via $P_4 = (P_2^2/2)(1 + \chi)$ to quantify the relative importance of stimulated emission. For a single-mode configuration $\chi$ is maximum and equal to 1. When multiple spatial and/or temporal modes are involved the visibility is reduced since the probability to create two pairs in the same mode is reduced. Only this contribution from pairs produced in the same mode is enhanced through stimulated emission. As a result, the visibility parameter $\chi$ is inversely proportional to the number of modes \cite{29,30}, and the number of spatial and temporal modes available in the down-conversion process are important quantities as they set an upper limit to the experimentally achievable visibility $\chi$.

4.2.1 Estimating the number of modes

Since the parametric down-conversion process generates either independent pairs, or exact copies of the pairs produced in the same mode the corresponding four-photon amplitude can be written as a product of two-photon amplitudes \cite{31} and a description in terms of a two-photon amplitude to find the corresponding number of modes suffices. This number of modes, or Schmidt number, involved in the parametric down-conversion process can be obtained via a Schmidt decomposition of the two-photon field. Estimates of the Schmidt number related to the spatial degrees of freedom are reported in literature for collinear \cite{22} as well as non-collinear \cite{23} geometries. Similarly, the effect of spectral filtering on the number of modes for pulsed PDC in a single spatial mode has been reported \cite{30,32}. Here we consider an experimental situation for crystals of different length where we include spatial as well as temporal modes. In this limit, analytical expressions can no longer be found. Instead of lengthy numerical computations we approximate the two-photon amplitude and use a two-photon coherence area to find approximate expressions that can be compared to the experiment.

The two-photon probability amplitude $C(q_s, q_i, \omega_s, \omega_i)$ for generating a non-collinear photon pair with transverse momenta $q_s$ and $q_i$ at frequencies $\omega_s$ and $\omega_i$ is given by

$$C(q_s, q_i, \omega_s, \omega_i) = E_p(q_s + q_i; \omega_s + \omega_i) \cdot T(\omega_s) \cdot T(\omega_i) \cdot \text{sinc}(\frac{1}{2} \Delta kL), \quad (4.1)$$

where $E_p(q_s + q_i; \omega_s + \omega_i)$ is the electric field amplitude of the pump pulse,
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and $T(\omega_s)$ and $T(\omega_i)$ are the transmission profiles of the spectral filters in the signal and idler beams, respectively. The term $\text{sinc}(\frac{1}{2} \Delta k L)$ is the phase-matching function. The phase mismatch $\Delta k L/2$ can be approximated using a Taylor expansion that includes the effect of index dispersion, and is given by

$$\frac{1}{2} \Delta k(\omega)L = b^2 |q_s - q_i|^2 + q_0 + \eta (\delta \omega_s + \delta \omega_i),$$

(4.2)

where the first two terms are the phase mismatch for the center wavelength with a collinear phase mismatch $q_0$, and parameter $b^2 = L/(4k_p)$, where $k_p = \omega_p n/c$ is the length of the wavevector at the center wavelength of the pump. The quantities $\delta \omega_s$ and $\delta \omega_i$ are the detuning of the angular frequency of the signal and idler frequency relative to the center frequency of the signal and idler beams. The phase mismatch $q_0$ controls the opening angle of the cone of down-converted light and can be tuned in the experiment through temperature tuning of the PPKTP crystal [33, 34].

The parameter $\eta = (DL)/2$ with $D = 1/v_g(\omega_p) - \frac{1}{2} (1/v_g(\omega_s) + 1/v_g(\omega_i))$ is the difference in inverse group velocities $v_g$ at the pump, signal and idler frequency [34, 35]. This term quantifies the difference in arrival time of pump and down-converted photons, and vanishes for small $\eta$ (small $D$ and/or $L$), where this walk-off is not important. We note that the group-delay dispersion $D$ as introduced here is only valid for type-I down-conversion where both down-converted photons have the same polarization [34, 35]. This is appropriate for the PPKTP crystals used in this study where pump and down-converted photons are all ordinarily polarized.

In order to address a more specific, experimentally relevant situation, and to allow for simple solutions, we introduce Gaussian approximations of the various factors in Eq. (4.1). We express the pump amplitude $E_p(q_s + q_i; \omega_s + \omega_i)$ as the product of two Gaussian functions

$$E_p(q_s + q_i; \omega_s + \omega_i) \propto \exp(-|q_s + q_i|^2/\sigma^2) \exp(-\tau^2(\delta \omega_s + \delta \omega_i)^2/2)$$

(4.3)

where $\sigma = 2/w_p$ is the inverse spatial width of the pump profile and $\tau = \tau_p/1.18$ characterizes the duration of the pump pulse, and $\tau_p$ is the duration of the pump pulse expressed as FWHM. We express the filter functions as $T(\omega_{s,i}) = \exp(-((\delta \omega_{s,i})/2F)^2)$, where $F$ is the filter bandwidth (FWHM), which is assumed to be much smaller than the natural bandwidth of the Type-I PDC process.
To approximate the phase-matching function by a Gaussian we use $\text{sinc}[\Delta k z/2] \approx \exp\left[-(1/4)(\Delta k L/2)^2\right]$. The phase-mismatch given by Eq. (4.2) shows that the spectral and spatial properties of PDC light are generally mixed. This implies that the color of the PDC light changes with emission angle. For colinear PDC this leads to non-separable ‘X’-entanglement [36] that becomes noticeable when a large frequency bandwidth of PDC light is collected. For non-colinear PDC, that we consider in this article, the correlations are transformed into a separable structure [37]. In addition, we collect only a small fraction of PDC light by using narrow frequency filters in the detection. Under those conditions the phase-matching function can be approximated as a separable product of two Gaussian functions to keep the analysis simple. The total two-photon probability amplitude of a pulsed pump can then be written as

$$C(q_s, q_i, \omega_s, \omega_i) \propto e^{-\langle \eta \rangle^2 (\delta \omega_s + \delta \omega_i)^2} \times e^{-(\nu^2/2)(\delta \omega_s + \delta \omega_i)^2} e^{-\langle 1/2F \rangle^2 (\delta \omega_s^2 + \delta \omega_i^2)} \times e^{-(\delta^2 |q_s - q_i|^2 + \phi_0)^2} e^{-(1/\sigma)^2 |q_s + q_i|^2}. \quad (4.4)$$

Despite the various approximations, the Schmidt decomposition of the two photon field of Eq. (4.4) has to be computed numerically. In this section we follow a different approach and estimate the visibility $\chi$ measured in the experiment as a function of the diameter of the pump beam, the length of the crystal, and the size of the aperture placed in the far field to collect the down-converted light.

Inspired by Eq. (4.4), we assume that the effect of the temporal modes, spatial modes and walk-off can be factorized so that their influence can be separated, i.e. $\chi = \chi_t \chi_s \chi_w$. This assumption is based on the fact that the spatio-temporal correlations of non-colinear PDC are separable and that second-order terms in the Taylor expansion of phase mismatch $\Delta k L/2$ are small. These terms give rise to additional mixing of spatial and temporal degrees of freedom and can be safely neglected for crystal lengths that are shorter than $\tau/D$ [37]. For the PPKTP crystals and the pulse duration in this study this length equals 1.3 mm and the neglecting the contribution of these terms is thus not strictly valid.

By treating the different degrees of freedom separately, we can use known expressions for the different visibilities. The visibility $\chi_t$, for PDC light filtered by a spectral filter with a bandwidth that is much more narrow than
the natural bandwidth of the process, is given by [30, 32]

$$\chi_t = \frac{1}{\sqrt{1 + (\tau F)^2}} \tag{4.5}$$

The influence of walk-off, characterized by the visibility $$\chi_w$$, can be estimated based on the two-photon field given by Eq. (4.4) and expressions found in literature for the Schmidt number of spatial modes for collinear [22] and non-collinear PDC [23] geometries. We write the dependence of the visibility on walk-off as

$$\chi_w = \frac{1}{\sqrt{1 + \left(\frac{L}{\pi a} \right)^2}}, \tag{4.6}$$

where $$L_0$$ is the total walk-off length as a result of both transverse and group-velocity walk-off.

The visibility $$\chi_s$$ can be determined by considering the correlations of two points at positions $$r_1$$ and $$r_2$$ in the far field, which are

$$\chi_s(r_1, r_2) = \exp(-|r_1 - r_2|^2 / r_0^2). \tag{4.7}$$

A coherence diameter $$r_0 = \lambda f / \pi w_p$$ has been introduced [29] as a measure for the diffraction limited momentum spread of the pump. Here $$\lambda$$ is the wavelength of the down-converted light and $$f$$ is the focal length of the lens used to create the far-field of the PDC source. To postulate these correlations, we have assumed that the coherence diameter $$r_0$$ is smaller than the width of the PDC ring. To keep a consistent notation we note that the quantity $$r_0$$ introduced here and the quantity $$\sigma$$ used in Eq. (4.4) are related via $$r_0 = 2\sigma f / k_p$$.

In the experiment, photons are collected through a finite aperture with a diameter $$a$$. Therefore the observed visibility of the four-photon state with two detectors is obtained by integration over the apertures, resulting in:

$$\chi_s(a) = \frac{1}{\pi^2 a^4} \int \chi_s(r_1, r_2) \Theta\left(\frac{a}{2} - |r_1|\right) \Theta\left(\frac{a}{2} - |r_2|\right) dr_1 dr_2, \tag{4.8}$$

where the Heaviside step-functions $$\Theta\left(\frac{a}{2} - |r_1|\right)$$ and $$\Theta\left(\frac{a}{2} - |r_2|\right)$$ represent the sharp edges of the two apertures with equal radius $$a/2$$ in the far-field. When both apertures are centered at the same position, the calculation of the visibility resembles that of efficient collection of two-photon states from a PDC source and the integral can be simplified to [38]:

$$\chi_s(a) = \frac{1}{\pi} \int_0^1 \left( \arccos(x) - x \sqrt{1 - x^2} \right) \exp\left(-\left(xa / r_0\right)^2\right) dx. \tag{4.9}$$
4.3 Experiment

The experimental setup to measure the visibility of the four-photon state is sketched in Fig. 4.1. UV pulses are produced by frequency doubling the pulses of a tunable mode locked Ti:sapphire laser operating at 826.4 nm wavelength. This results in ~ 2 ps long pulses at 80 MHz repetition rate, with approximately 180 mW average laser power at a wavelength of 413.2 nm. Lens $L_1$ focuses the pulsed laser beam to a diameter $w_p$ into a periodically poled KTP (PPKTP) crystal that generates non-collinear, frequency degenerate photon pairs at a wavelength of 826.4 nm [34]. To this end the PPKTP crystal is temperature controlled, and stabilized within 20 mK, to adjust the phase-matching conditions. Typically, we set a constant temperature of $T = 25^\circ C$ of the PPKTP crystal to create an opening angle $\theta \approx 40$ mrad for the down-converted light.

The PDC light is collected by lens $L_2$ with a 270 mm focal distance and is filtered by a bandpass filter $F$ with a bandwidth of either 1.0 nm or 0.4 nm FWHM. The photons are split by a beamsplitter $BS$ and are then spatially filtered by the apertures $A$ and collected into a 50 $\mu$m core multi-mode fiber using lens $L_3$ ($f_3 = 11$ mm). The photons are then detected by fiber-coupled single photon counting modules. The combination of aperture, lens and fiber acts as a bucket detector and is mounted on a motorized translation stage to scan the detection unit in $x$ and $y$-directions in the far field of the PDC source created by the lens $L_2$. In the experiment we convert the position of the detector in the far-field to an angle by dividing by the focal length of the lens. This angle, denoted as $q$, corresponds to the transverse momentum $q$ normalized to the wavevector of the PDC light, i.e. $q = 2|q|/k_p$.

An electronic time delay of either 0 ns or 12 ns is introduced between detectors $D_1$ and $D_2$ to record coincidence events in the same pulse ($R_{0\text{ns}}$) and between subsequent pulses ($R_{12\text{ns}}$). For count rates small compared to the laser repetition rate the contribution from more than two pairs to the coincidence rates can be neglected. The coincidence rate $R_{12\text{ns}}$ is due to two independent pairs produced in the two different laser pulses, and is given by

$$R_{12\text{ns}} = N_P \eta^2 \frac{P_2^2}{2},$$

where $N_P$ is the laser repetition rate, $P_2$ is the probability to create a single photon pair via spontaneous PDC, and $\eta/2$ is the photon detection efficiency.
4.3 Experiment

Figure 4.1: Experimental setup for generating and characterizing spatially entangled four-photon states. UV pump pulses from a frequency doubled Ti:Sapphire laser are focused by lens $L_1$ in a PPKTP crystal. Down-converted photons are collected by the lens $L_2$ ($f_2 = 270$ mm) and filtered by a bandpass filters $F$ at a wavelength of 826.4 nm with 1 nm or 0.4 nm FWHM bandwidths. Photons are split by a beam splitter (BS) and detected by fiber-coupled APDs $D_1$ and $D_2$ placed on computer controlled translation stages. Spatial-mode selection occurs by means of tunable detector apertures (A) and a lens $L_3$ mounted on a translation stage. Photon counts and coincidences are recorded as a function of the angular positions $q$ (horizontal direction) and $p$ (vertical direction) of the detectors in the far-field.

that contains an extra factor two to accounts for the beamsplitter. Similarly, the coincidence count rate $R^{0ns}$ contains a contribution due to stimulated emission and is given by

$$R^{0ns} = N_P\left(\frac{\eta}{2}\right)^2 \frac{P_2}{2} (1 + \chi),$$

(4.11)

In the experiment we find the visibility parameter as

$$\chi = \frac{(R^{0ns} - R^{12ns})}{R^{12ns}} \quad [29],$$

assuming that contributions to the coincidence rate from quantum states with more than 4 photons remain small in the experiment. Since the setup collects photons from one side of the PDC ring, two signal photons created by stimulated emission (the corresponding two idler photons are located on the other side of the ring) are split on the beam splitter and detected in the far field by detectors $D_1$ and $D_2$. Only
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photon states produced by stimulated emission give rise to the correlations that register as coincidences. Hence, this setup with two detectors suffices to monitor the correlations of a four-photon state [29, 30].

4.4 Results and Discussion

4.4.1 Four-photon visibility in the spatial and temporal domain

Figure 4.2 shows the measured visibility of the four-photon state as a function of aperture size for a 2 mm long PPKTP crystal and a pump beam focused to a waist $w_p = 85 \pm 10 \mu m$. Measurements are shown for a 1 nm and 0.4 nm FWHM bandpass filter. The transmission spectra of the two bandpass filters are shown in the inset. The different symbols in the figure correspond to different measurement series. As can be seen, the visibility increases for smaller aperture size and more narrow bandpass filters. The solid lines through the data are calculations based on numerical integration of Eq. (4.8) using a fixed coherence diameter $r_0 = 0.84 \pm 0.1$ mm, which is calculated from the independently measured beam waist. The shaded areas in Fig. 4.2 show the outcome of similar calculations based on a realistic range of beam diameters of $w_p = 75 - 95 \mu m$ in the experiment. Spectral filtering with the 0.4 nm FWHM bandpass filter yields a twice higher visibility ($\chi_t = 0.8$) than filtering with the 1 nm FWHM bandpass filter, consistent with the dependence of $\chi_t$ on filter bandwidth.

4.4.2 Universal expression for visibility

The far-field size of a spatial mode, and thus the coherence area $r_0$, increases for tighter focusing conditions. The diameter and width of the PDC ring remain constant as long as the Rayleigh range of the focused beam is significantly larger than the crystal length. As a consequence, an area on the PDC ring selected by an aperture with diameter $a$ in the far field is expected to yield a higher visibility of four-photon states when created with a tighter focus.

The measured visibility of the four-photon states as a function of aperture size for three different focusing conditions ($w_p = 55 \pm 5 \mu m$ (black symbols), $w_p = 85 \pm 5 \mu m$ (red symbols) and $w_p = 155 \pm 5 \mu m$ (blue symbols)) is shown in Fig. 4.3(a), where the different symbols refer to different measure-
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Figure 4.2: Variation of the visibility $\chi$ of the four-photon state as a function of aperture size. The PDC light is generated in a 2 mm PPKTP crystal and detected using 1 nm FWHM bandpass filter (black open symbols) and 0.4 nm FWHM bandpass filter in combination with the 1 nm FWHM filter (red solid symbols) at 826.4 nm wavelength. The solid lines (with shaded areas) correspond to calculations based on Eq. (4.8) with pump beam diameter of $85\pm10 \, \mu m$ for each of the bandpass filters. The inset shows the measured transmission spectra of the filters.

The solid lines in Fig. 4.3(a) indicate the calculated visibility as a function of detector aperture size $a$ obtained by numerical evaluation of Eq. (4.8). This calculation contains no free parameters other than the number of temporal modes, determined by the 1 nm bandpass filter, which limits the maximum visibility to $\chi_0=0.4$. The value of $r_0$ calculated from the measured beam diameter for the different focussing conditions is $r_0 = 1.42 \pm 0.14 \, \text{mm}$, $0.84 \pm 0.08 \, \text{mm}$ and $0.46 \pm 0.05 \, \text{mm}$ for a pump beam waist of $55\pm5$, $85\pm5$ and $155\pm5 \, \mu m$, respectively. As can be seen, the model is in excellent agreement with the experimental data.

The similarity of the curves in Fig. 4.3(a) and the prominent role of $r_0$
Figure 4.3: (a) Visibility $\chi$ of four-photon states obtained using different pump beam diameters equal to $w_p = 55 \pm 10 \, \mu m$ (black symbols), $w_p = 85 \pm 10 \, \mu m$ (red symbols) and $w_p = 155 \pm 10 \, \mu m$ (blue symbols) as a function of the diameter of the detector aperture. Photon pairs are created in a 2 mm long PPKTP crystal and filtered by a 1 nm FWHM bandpass filter. (b) Universal curve for visibility of four-photon states as a function of the normalized aperture diameter $a/r_0$. The inset illustrates one side of the PDC ring with an area selected by an aperture of diameter $a$ together with the characteristic diameter $r_0 = \lambda f / (\pi w_p)$ determined by the diameter of the pump beam $w_p$.

suggest the existence of a universal relationship for the four-photon visibility. This universal curve is expressed in integral form by Eq. (4.9), which contains the ratio $a/r_0$. Figure 4.3(b) shows the same data rescaled by plotting the visibility as a function of the ratio of aperture and coherence diameter $a/r_0$. 
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All measurements now fall onto the same curve for the visibility as a function of the dimensionless parameter $a/r_0$. We note that the universal curve in Fig. 4.3(b) can only be valid as long as the detector aperture is smaller than the width of the PDC ring (see the inset of Fig. 4.3(b)), so that the coherence expressed by Eq. (4.8) remains independent of the width of the PDC ring. Hence, for a given combination of aperture size and pump beam diameter only part of the curve can be recovered.

4.4.3 Spatial and temporal walk-off

The measured visibility also depends on the length of the crystal through walk-off. This walk-off makes the stimulated emission process less probable since different sections of the crystal function as independent sources. The corresponding visibility of the four-photon states is reduced and should be inversely proportional to the crystal length for crystals that are much longer than the walk-off length.

We limit the discussion to a transverse walk-off and a transit-time difference walk-off because birefringent walk-off is not present in this study that uses a PDC source based on PPKTP crystals where all photons have the same polarization. The transit-time difference walk-off is related to the transit-time dispersion $D = 1.5 \text{ ps/mm}$ [35] and puts a limit to the crystal length over which the down conversion is efficient [34]. This walk-off becomes important when the delay between pump and down-converted pulses is comparable to the pulse duration $\tau$. The corresponding group velocity walk-off length is defined as $L_g = \tau/D$. For the $\sim 2$ ps laser pulses used here we estimate a walk-off length 1.3 mm.

For non-collinear PDC, an additional transverse walk-off is present because the down-converted photon beams propagate at an angle relative to the pump beam. The transverse walk-off length is defined via $L_t = w_p n/\theta_0$, where $w_p$ is the beam diameter of the pump beam, $n$ is the (ordinary) refractive index of the crystal and $\theta_0 \approx 40$ mrad is the external opening angle of the PDC light. The corresponding transverse walk-off length for a realistic beam waist of 100 $\mu$m is $L_t \approx 4.3$ mm, i.e. a factor 3 larger than the group-velocity walk-off length $L_g$.

Figure 4.4 shows the measured visibility as a function of crystal length for three different focussing conditions obtained by measuring the extra co-
Figure 4.4: Measured visibility $\chi$ of the four-photon state generated with pump beam diameters equal to $w_p = 55 \, \mu m$ (blue triangles), $w_p = 85 \, \mu m$ (red circles) and $w_p = 155 \, \mu m$ (green squares). The data are shown as a function of crystal length, using an aperture diameter of 1 mm and a 1 nm FWHM bandpass filter. The solid lines represent fits to the data (see text). The shaded areas indicate the confidence interval of the fit taking into account a $\pm 10 \, \mu m$ uncertainty in the beam diameter $w_p$. The inset shows the walk-off length obtained from the fit of Eq. (4.6) to the data.

incidence events for crystal lengths of 2, 5, 10 and 20 mm. The solid lines in Fig. 4.4 correspond to a best fit of the visibility $\chi$ given by Eq. (4.6) with only $L_0$ as a fit parameter to the data. The shaded areas indicate a range of realistic fit curves incorporating a range of beam diameters based on a $\pm 10 \, \mu m$ uncertainty in the measured beam diameter. In the fit procedure, we reduce the number of fit parameters by considering the contribution to the visibility due to walk-off $\chi_w$, temporal modes $\chi_t$ and spatial modes $\chi_s$ independently, i.e. by using $\chi = \chi_w \chi_t \chi_s$. The value of $\chi_t = 0.4$ follows from the 1.0 nm spectral bandwidth of the filter used. The value of $\chi_s$ can be estimated from Fig. 4.3 using the known aperture diameter $a = 1$ mm and the value of $w_p$. We estimate $\chi_s = (1/1.03), (1/1.23)$ and $(1/2.07)$ for $w_p = 55 \, \mu m$, 85 $\mu m$, 155 $\mu m$, respectively.

All measurements are in good agreement with the model given by
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Eq. (4.6) and can be described by a walk-off length \( L_0 \approx 1.0 \pm 0.1 \) mm that is comparable to the group-velocity walk-off length \( L_g \) and is independent of the pump beam waist \( w_p \). The inset of Fig. 4.4 shows the walk-off length as a function of the pump beam waist obtained from a best fit with error bars as a result of experimental uncertainty in beam diameter. These data suggests a trend of decreasing walk-off length \( L_0 \) with beam diameter, which is surprising as we would expect a weak increase of the walk-off length with beam diameter because spatial walk-off becomes less important for larger beam diameters. We suggest that the reason for this deviation are terms that mix temporal and spatial degrees of freedom in the two-photon amplitude. These correlations between the frequency of PDC light and transverse momentum will indeed become more apparent in the experiment for smaller \( r_0 \), i.e. for larger pump beam diameters.

4.4.4 Joint spatial distribution of four-photon states

The model for the spatial visibility \( \chi_s \) as expressed by Eqns. 4.7 and 4.8 is valid when the intensity of the down-converted source can be assumed constant over the PDC ring. Strictly speaking this condition is difficult to achieve in an experiment since the sizes of the aperture \( a \), the parameter \( r_0 \) as well as the width of the PDC ring in the far field are of the same order of magnitude. It is thus rather surprising that the model works so well and that a universal curve as a function of dimensionless aperture size describes our data. This raises questions how far the visibility model with a simple coherence area can be used outside the range for which it was designed and what are the features of experiments where this model starts to break down.

To investigate this question and to probe the limits of the validity of the model, we measure the coincidence rates as a function of the angular position (transverse momentum in the horizontal (x) direction) \((q_1, q_2)\) of the two apertures in the far field created by lens \( L_2 \), for different focussing conditions. From these measurements the excess coincidence rate as \((R^{0ns} - R^{12ns})\) can be determined. Compared to the visibility this quantity is easier to measure for points at the edge of the PDC ring with a low coincidence rate, and we refer to this measurement as a joint spatial distribution of stimulated pair emission [29]. To convert these values to a visibility the rate should be divided by the rate \( R^{12ns} \).
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Figure 4.5: Measured joint spatial distribution of genuine four-photon states as a function of the angular positions of \( q_1 \) and \( q_2 \) of the detectors \( D_1 \) and \( D_2 \) in the far-field using a 1 nm FWHM bandpass filter and a 1.5 mm aperture size at \( f = 270 \) mm. Down-converted photons are created by a pump beam diameter of (a) \( w_p = 35 \pm 5 \) \( \mu \)m (b) \( w_p = 45 \pm 5 \) \( \mu \)m (c) \( w_p = 85 \pm 5 \) \( \mu \m). The false color images in Fig. 4.5 show the measured excess coincidences for a pump beam diameter equal to \( w_p = 35 \pm 5 \) \( \mu \)m (a), \( w_p = 45 \pm 5 \) \( \mu \)m (b) and \( w_p = 80 \pm 5 \) \( \mu \m \) (c). Data are represented as a function of the angular position \( q_1 \) and \( q_2 \) of the two scanning detectors \( D_1 \) and \( D_2 \). To perform these measurements a spectral filter with a 1.0 nm bandwidth and an aperture with a diameter \( a = 1.5 \) mm are chosen as a compromise between getting a sufficiently high visibility and a reasonable coincidence count rate. This allows to measure the complete spatial correlations within 4 hours using a point-by-point scan, before realignment of the setup is necessary.

The data in Fig. 4.5 clearly shows an ellipsoidal feature in the excess coincidences with the long axis aligned along the diagonal direction. This diagonal direction corresponds to two detectors looking at the same position in the far field of the PDC source, where strong correlation produced by a stimulated pair emission are expected. To compare the measurements to the model expressed by Eq. (4.8), we need to normalize the measured difference in coincidence rate by the accidental rate \( R^{12ns} \). To this end, we fit both the corrected coincidence rate and the accidental coincidence rate to a 2-D Gaussian distribution. These fits yield characteristic sizes \( r_1 \) and \( r_2 \) for the excess coincidences, as indicated in Fig. 4.5(a), and a single size \( r_a \) for the accidental
rate. We use the fitted values of \( r_1, r_2 \) and \( r_a \) to compute the size of the correlations in the visibility. The spatial visibility \( \chi_s \) is also ellipsoidal, with sizes \( r'_1 \) and \( r'_2 \). These sizes are defined via \( \left( \frac{1}{r_{12}} \right)^2 = \left( \frac{1}{r'_1} \right)^2 + \left( \frac{1}{r_a} \right)^2 \) and are larger than the sizes visible in Fig. 4.5.

**Table 4.1:** The experimental and calculated values of characteristic diameter \( r'_1 \) and the visibility \( \chi_t \chi_s \) (see text).

| \( w_p (\mu m) \) | \( r_0 (\text{mrad}) \) | \( r'_1 (\text{mrad}) \) | \( \chi_t \chi_s \) | \( r'_1 (\text{mrad}) \) | \( \chi_t \chi_s \) |
|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| 35 ± 5            | 7.5 ± 1.1         | 11.2 ± 0.5        | 0.21 ± 0.08       | 9.4 ± 1.3         | 0.35 ± 0.02       |
| 45 ± 5            | 5.8 ± 0.6         | 7.8 ± 0.3         | 0.31 ± 0.08       | 7.6 ± 0.7         | 0.32 ± 0.02       |
| 85 ± 5            | 3.3 ± 0.2         | 5.8 ± 0.3         | 0.22 ± 0.05       | 5.2 ± 0.2         | 0.23 ± 0.01       |

Table 4.1 summarizes the values of \( r'_1 \) as well as the visibility \( \chi_t \chi_s \) deduced from the experiment for the different pump beam waists. The value of \( r_0 \) is included as a reference. The error bars on \( r'_1 \) follow from error propagation using the fitted values \( r_1 \) and \( r_a \). The value of \( \chi_t \chi_s \) in the experiment was determined by calculating the average value along the diagonal in Fig. 4.5. The experimental values should be compared to a theoretical expression, similar to Eq. (4.8) modified so that the integration runs over the two apertures that are no longer centered at the origin. This model predicts that the visibility is independent of \( q_1 + q_2 \) and therefore predicts \( r'_2 \) to be infinite. This is consistent with the data for the 85 \( \mu m \) pump beam size where the width of the accidental coincidences and the extra coincidences is identical within the error bar. For tighter focusing conditions the value of \( r'_2 \) deduced from the experiment is finite.

The calculated values for the visibility \( \chi_t \chi_s \) and \( r'_1 \) are summarized in Table 4.1, where the error bars are calculated taking into account the uncertainty on the measured pump beam waist. Good agreement with the experiment is found for the 45 and 80 \( \mu m \) pump beam waist. For the strongest pump beam focus, the calculated visibility and size \( r'_1 \) do not agree with the experiment. Our interpretation is that the assumptions underpinning the model of Eq. (4.8) break down. For the strongest focusing conditions, the spread in wavevector of the pump affects the phase-matching conditions as the crystal
length becomes comparable to the Rayleigh range of the beam. This leads to a PDC ring that is broadened in the far-field due to the focussed pump beam. Using the notation of Law and Eberly [22], this corresponds to the regime where $b \sigma > 1$. In this regime the number of Schmidt modes increases and the visibility $\chi_s$ is reduced. This decrease in visibility for very tight focussing is not contained in Eqns. 4.7 and 4.8, that predict a monotonous increase with decreasing pump-beam waist, i.e. an increase in the far-field mode size $r_0$.

4.5 Conclusion

We have characterized four-photon spatial correlations created via stimulated parametric down-conversion in a non-collinear geometry. High visibility of the four-photon state up to 80% is observed by narrow spectral and spatial filtering of the down-converted light. This demonstrates that single (spatial and temporal) mode operation can be achieved under suitable experimental conditions.

Data for different focussing conditions can be combined to a single, universal expression for the visibility of the four-photon state. This universal expression does not depend on material properties and contains only the ratio of the aperture size $a$ and a spatial coherence diameter $r_0$ set by the divergence of the pump beam. Our experimental results are consistent with this universal expression of the visibility. By varying the size $r_0$ using different focusing conditions we ensure that the aperture size $a$ remains smaller than the width of the PDC ring. This allows to explore the expression for visibility over a significantly larger range of the parameter $a/r_0$ than what is possible in a single experiment.

For longer crystals pumped by a picosecond pulsed pump laser, the combination of the crystal length and opening angle of the PDC cone introduces a spatio-temporal walk-off effect. We have explored this effect by measuring the visibility for different focussing conditions as a function of crystal length. The experimental data can be described by a walk-off length of $1.0 \pm 0.1$ mm for a 2 ps long pulse. This walk-off length is comparable to the group velocity walk-off length, estimated to be 1.3 mm. Transverse walk-off effects were found to be relatively unimportant.

The detailed structure of the spatial correlations expressed as a joint spatial distribution of the four-photon state show that the model and the uni-
versal curve break down for tight focussing. The tight-focussing condition mixes the spatial and temporal degrees of freedom. For a 2 mm long PPKTP crystal, this becomes apparent for a laser beam waist smaller than 40 µm.

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