Photon pair generation using four-wave mixing in a microstructured fibre: theory versus experiment

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\textbf{Abstract.} We develop a theoretical analysis of four-wave mixing used to generate photon pairs useful for quantum information processing. The analysis applies to a single mode microstructured fibre pumped by an ultra-short coherent pulse in the normal dispersion region. Given the values of the optical propagation constant inside the fibre, we can estimate the created number of photon pairs per pulse, their central wavelength and their respective bandwidth. We use the experimental results from a picosecond source of correlated photon pairs using a micro-structured fibre to validate the model. The fibre is pumped in the normal dispersion regime at 708 nm and phase matching is satisfied for widely spaced parametric wavelengths of 586 and 894 nm. We measure the number of photons per pulse using a loss-independent coincidence scheme and compare the results with the theoretical expectation. We show a good agreement between the theoretical expectations and the experimental results for various fibre lengths and pump powers.
1. Introduction

Quantum information can be encoded in various media including atoms, ions and electrons. It is photons, however, that are the most useful for transporting quantum information between separate locations. They are often called flying Q-bits and they appear to be the fundamental resource for quantum communications experiments [1, 2]. Their robustness to decoherence also leads them to be involved in various multiphoton and linear optical logic experiments [3]–[5]. One of the main resources needed for these quantum optics experiments is the ability to create these photons in pairs. When simply correlated in time they can be used to create heralded single photons [6, 7] (closely approximating true single photon sources). More interesting is when these photons have strongly correlated properties; they are entangled [8, 9]. Such sources continue to be exploited in fundamental multiphoton experiments such as quantum teleportation [10] or to create cluster states [11]–[13].

The preferred sources for such experiments until recently have been three-wave mixing in $\chi^{(2)}$ nonlinear birefringent crystals [8, 9]. These sources are inherently wide band, low brightness (per nanometre, per single mode) sources. The increasing number of photons involved in current experiments is requiring brighter sources and narrower photon bandwidths. Hence, periodically poled fibres [14] and periodically poled waveguides of lithium niobate [15] have been shown to be bright pair photon sources. In poled fibres [16] the low nonlinearity limits the brightness while in planar waveguides the non-circular mode limits the coupling efficiency into optical fibres [17].

It is well known that parametric gain can arise from the $\chi^{(3)}$ nonlinearity in optical fibres [18, 19] and phase matching can be achieved by using the modulation instability when pumping...
fibres in their anomalous dispersion regime. Various pair photon generation experiments have been performed in this regime [20]–[24]. The photon pairs are generated close to the pump wavelength and are always accompanied by a significant Raman background and careful filtering is required. We have shown that phase matching can be obtained for widely spaced wavelengths by pumping photonic crystal fibre (PCF) close to the zero dispersion wavelength in the normal dispersion regime [25]–[28]. Recently, we have used a fibre with the zero dispersion point in the near infrared (715 nm) and pumped with a picosecond pulsed laser, red detuned a few nanometres into the normal dispersion regime, to demonstrate a high brightness source of photons pairs [29].

We are seeking to develop a source which may be applicable for future quantum interference experiments involving three or more photons created as two or more pairs. Interference effects between separate pairs of photons can be studied by overlapping photons with a time uncertainty shorter than their inverse bandwidth or coherence length [30]. This restricts us to sources pumped by ultra-short laser pulses where the bandwidth is of the order of nanometres and also requires a high efficiency of collection. Theoretical models of correlated photon pair generation via four-wave mixing (FWM) have already been investigated [19, 31]. The reference [19] is restricted to monochromatic pump with infinite coherence length. It does not apply to pulsed experiments. The second reference [31] is the first to investigate the influence of the finite pump bandwidth of narrow pulses in FWM. We propose here a theory that goes further in the calculation and develop a model that, given the fibre geometry and its dispersion properties, predicts the number of expected photons and their spectral properties from a quantitative point of view without fitting parameters.

We rigorously compare our model to experimental results from correlated photon pairs generated in a PCF. The paper is structured along the following lines: section 2 presents the quantum theory of FWM in optical fibres, while section 3 is dedicated at tagging experimental parameters to the theory. Section 4 describes and analyses the experimental results from our PCF. We investigate in this section the agreement between theoretical expectations and experimental results. Section 5 outlines the possibilities of using this fibre as a source of multiphoton pairs for quantum information processing. Finally, we briefly conclude in section 6.

2. FWM process in fibre with pulsed pumping

2.1. Definition of the involved fields

We are looking at the process of FWM based on the nonlinear susceptibility $\chi^{(3)}$ in a PCF where two pump-photons are converted into a pair of signal and idler photons. We are specializing to energy matched situations where $2\Omega_p = \omega_s + \omega_i$. The related interaction Hamiltonian in a volume within the fibre is:

$$H_{\text{int}} = \int_V U \, dV$$

(1)

and the energy density associated with four fields coupled in a third-order nonlinear medium is:

$$U = \varepsilon_0 \chi^{(3)}(\Omega_p, \omega_s, \omega_i) E_p^2 E_s E_i.$$  

(2)
Here $\epsilon_0$ is the permittivity of free space allowing us to measure $\chi^{(3)}$ in units of $V^{-2} \text{m}^2$. In the fibre optic community, it is common to rather use the nonlinear refractive index $n_2$ defined as follow:

$$n_2 = \frac{3\chi^{(3)}}{4\epsilon_0 cn_0^2},$$

(3)

where $n_0$ is the refractive index of the fibre.

For a non-stationary pumping pulse, the field amplitude can be decomposed into its Fourier components in the following form:

$$E_p(x, y, z, t) = \int_{-\infty}^{+\infty} E_p(\omega, x, y, z, t) \, d\omega.$$  

(4)

In our guided configuration all the involved fields are propagating in the same direction, hence we insert $e_p(\vec{r})$ as the transverse spatial variation with normalization $\int |e_p(\vec{r})|^2 \, dr \, d\theta = 1$ inside all our equation and carry out the calculation along the $Z$-axis which is the fibre axis. We assume that $E_p(x, y, z, t)$ is a strong classical Gaussian pump beam of full-width half-maximum (FWHM) bandwidth $\Delta \omega_p$ and central frequency $\Omega_p$. We focus on one of its Fourier component defined as follows:

$$E_p(\omega) = \frac{E_p}{2} G_p(\omega) \left( e^{-\omega^2 / 2\sigma^2} + e^{i(\Omega_p + \omega) t - (k_p - \gamma P_p)z} \right) e_p(\vec{r}),$$  

(5)

where $\gamma P_p$ is the self-phase modulation coefficient acquired by the pump pulse as it propagates within the fibre; $G_p(\omega) = e^{-\omega^2 / 2\sigma^2}$ is the weight of the component $\Omega_p + \omega$ of the pulse and $1 / \sigma^2$ is the variance characterizing the pump pulse. The variance is linked to the bandwidth of the pulse by $\Delta \omega_p = 2\sqrt{\ln(4)} / \sigma$. We define the energy density within a pulse so that (Parseval’s theorem):

$$U_{\text{pulse}} = \epsilon_0 \int_{-\infty}^{+\infty} E_p(\omega) E_p^*(\omega) \, d\omega \approx \epsilon_0 \int_{-\infty}^{+\infty} \left| E_p e^{-\omega^2 / 2\sigma^2} \right|^2 \, d\omega.$$  

(6)

While the strong pump pulse remains classical, here the two-photon modes associated with $E_s$ and $E_i$ are quantized using the creation and annihilation operators and we use similar spatial terms $e_l(\vec{r})$ to describe their transverse variation with normalization $\int |e_l(\vec{r})|^2 \, dr \, d\theta = 1$.

$$E_l = \sqrt{\frac{\hbar \omega_l}{2\epsilon_l}} \frac{1}{\sqrt{\mathcal{L}^3}} \sum_{k_i} (a_{i}^{\dagger} e^{-ik_i z} - a_{i} e^{ik_i z}) e_l(\vec{r}),$$  

(7)

where $\mathcal{L}^3$ defines the quantization volume.

2.2. Interaction Hamiltonian

We calculate the interaction Hamiltonian for one monochromatic component of the pump pulse and will sum up over all the possible combinations between the components at a later stage.
Starting with the interaction Hamiltonian for the parametric process, we can easily derive the interaction Hamiltonian for two pump-components $\omega_{p1}$ and $\omega_{p2}$ as:

$$H_{\text{int}} = S I \hbar \sum_{k_s,k_i} G_p(\omega_{p1}) G_p(\omega_{p2}) \left[ a_s^\dagger a_i^\dagger e^{-i(2\Omega_p+\omega_{p1}+\omega_{p2})t} L \text{sinc} \left( \frac{\Delta k L}{2} \right) + \text{h.c.} \right],$$  \hspace{1cm} (8)$$

where $I = \iint e_{p1}(\vec{r}) e_{p2}(\vec{r}) e_s(\vec{r}) e_i(\vec{r}) \, d\vec{r} \, d\theta$ is a normalized factor; $\Delta k = k_{p1} + k_{p2} - k_s - k_i - 2\gamma P_p$ is the phase-matching coefficient and $S$ is the gain parameter defined as follows:

$$S = \epsilon_0 \chi^{(3)} \frac{E_{p0}^2}{4} \sqrt{\frac{\omega_s \omega_i}{4 \epsilon_s \epsilon_i}}.$$  \hspace{1cm} (9)$$

The sinc function in (8) gives rise to the phase-matching condition between the pump components, signal and idler fields. The energy associated with the individual signal and idler fields in the fibre is defined by

$$H_0 = \hbar \left[ \omega_s (a_s^\dagger a_s + \frac{1}{2}) + \omega_i (a_i^\dagger a_i + \frac{1}{2}) \right]$$  \hspace{1cm} (10)$$

leading to a total Hamiltonian to be:

$$H_{\text{tot}} = H_o + H_{\text{int}}.$$  \hspace{1cm} (11)$$

Remembering that this Hamiltonian applies only for two arbitrary monochromatic pump components, we will later integrate over the full bandwidth of the pump pulse.

2.3. The Heisenberg equation of motion

In the standard Heisenberg representation the evolution of any operator $A$ is defined by

$$\frac{dA}{dt} = -\frac{i}{\hbar} \left[ A, H_{\text{tot}} \right].$$  \hspace{1cm} (12)$$

In the case of $A = a_s^\dagger$ and assuming $a_i(t) = \tilde{a}_i(t)e^{-i\omega_it}$, equation (12) gives:

$$\tilde{a}_s^\dagger(t \to \infty) = \tilde{a}_s^\dagger(0) - \frac{S I L}{2\sqrt{\mathcal{L}}} \sum_{k} G_p(\omega_{p1}) G_p(\omega_{p2}) \tilde{a}_i(0) \delta(\Delta\omega) \text{sinc} \left( \frac{\Delta k L}{2} \right).$$  \hspace{1cm} (13)$$

When integrating to obtain (13) we assumed that the system (nonlinear medium and the pump components) is turned on at time zero and runs continuously. This gave rise to the $\delta$-function in $\Delta\omega$ for the frequencies of the fields to guarantee the energy conservation at single-photon level for a monochromatic pump component.

We then perform the integration over all the possible combinations of $\omega_{p1}$ and $\omega_{p2}$ within the pump bandwidth. Basically if we define the interaction satisfying the basic energy conservation $2\Omega_p = \omega_s + \omega_i$ and introduce $\Delta\omega_{k_s}$ as the frequency shift for the signal mode $k_s$ compared to
the central one $\omega_{s}$ (respectively $\Delta \omega_{k_i}$ for the idler mode $k_i$), the $\delta$-function then helps to carry out the double integral over $\{\omega_{p_1}, \omega_{p_2}\}$ and for $\tilde{a}_s(\infty)$ we get

$$\tilde{a}_s(\infty) = \tilde{a}_s(0) - \frac{\text{SIL}}{2} \sqrt{\frac{\pi}{\sigma}} \sum_{k_i} e^{-\frac{(\Delta \omega_{k_i} + \Delta \omega_{k_s})^2}{2\sigma^2}} \tilde{a}_i(0) \text{sinc} \left( \frac{\Delta k L}{2} \right).$$

(14)

It is interesting to note that in (14) the dirac function (standing for the energy conservation in monochromatic pump regime) turns here into an exponential function guaranteeing the energy conservation in the broadband pump regime.

In order to calculate the mean number of photons $\langle N_s \rangle$ over all the possible modes $k_s$, we define the operator:

$$W_s(t) = \sqrt{\frac{1}{L}} \sum_{k_s} \tilde{a}_s(t).$$

In the limit $L \to \infty$, the sum over $k_{s,i}$ ($\frac{1}{L} \sum_{k} e^{-\frac{\Delta \omega_{k}^2}{\sigma^2}}$) can be replaced by an integral ($\int dk$) in the usual way so that we finally have:

$$\langle N_s \rangle = \int \langle 0 \vert W_s^\dagger W_s \vert 0 \rangle dt$$

$$= \left( \frac{\text{SIL}}{2} \right)^2 \sqrt{\frac{\pi}{\sigma}} \int \int \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) e^{-\frac{(\Delta \omega_{k_s} + \Delta \omega_{k_i})^2}{2\sigma^2}} \frac{d\Delta \omega_{k_s}}{v_{g_s}} \frac{d\Delta \omega_{k_i}}{v_{g_i}},$$

(15)

where $v_{g_s}$ and $v_{g_i}$ are the group velocity of signal and idler wave in the fibre.

We show in the following section that (15) becomes easy to integrate under certain approximations and we derive the expected mean number of photon per pulse at the output of our PCF.

3. From theory to experiment

Figure 1 shows the microstructured fibre used in our experiment. The fibre has a core diameter of 2 $\mu$m and presents a zero dispersion wavelength $\lambda_0 = 715$ nm. Typical spectra obtained when pumping with 708.4 nm light and aligning the polarization on one of the axes of the fibre are shown in figure 2. The asymmetry of the microstructure makes the fibre slightly birefringent. As a consequence, when we do not launch into the correct fibre polarization axis, we see two peaks corresponding to different phase-matching conditions for the different fibre axes. The absence of the second peak here implies our source is strongly polarized. The short wavelength sits at 586.4 nm and the corresponding idler at 893.9 nm. They feature an associated FWHM bandwidth of 4.5, 0.4 and 9.6 nm. The spectra were taken at a pump power of 1 mW and flat background comes entirely from the electronic bias errors in the CCD. Then by tuning the pump photon wavelength and monitoring similar spectra, we are able to observe the evolution of the signal and idler wavelength versus the pump wavelength (see figure 3). These wavelengths are those which satisfy the following energy conservation:

$$2\Omega_p - \omega_s - \omega_i = 0$$

(16)
Figure 1. Electron microscope image of the PCF used with core diameter $d \approx 2 \mu m$ and $\lambda_0 = 715$ nm.

Figure 2. Fluorescence spectrum of the signal, pump and idler photons over 10 s. The number of counts is proportional to the number of photons detected by the cooled camera. The small peak at 598 nm is attributed to unwanted background light or a detector fault since it is visible even when the laser is blocked.

and phase-matching equations

$$2 \frac{n_p \Omega_p}{c} - \frac{n_s \omega_s}{c} - \frac{n_i \omega_i}{c} - 2 \gamma P = 0,$$

where $n_{p,s,i}$ are the refractive index of the medium at pump, signal and idler wavelengths. The phase shift acquired by the pump waves (self-phase modulation) is defined by the factor $\gamma P$ [18]. We also present on figure 3, the theoretical phase-matched curve using the propagation constant for a simple strand of silica in air as a close approximation [32] to the fibre used in this experiment. This approximation does not give a good agreement between the theoretical expectations and the experimental results as it does not take into account the role played by the honeycomb structure.
3.1. Phase-matched solutions

As the percentage of silica in the cladding is not negligible, the accuracy of this model is not perfect. To solve this problem we use a novel method [33] to determine the propagation constant of a PCF. This method models the PCF as a step-index fibre with a core index set to that of fused silica, and a cladding index set to the mean index of the PCF’s cladding. We used an effective cladding index of $n = 1.05$ (corresponding to 90% air-filling fraction) to retrieve the propagation constant of the fibre and obtain the fit shown in figure 3. There is a very good agreement, confirming that our core diameter is actually 2 $\mu$m and that the honeycomb structure does contribute to the effective refractive index of the fibre cladding, perturbing the propagation constant from that generated by the naked strand model.

We have now a good knowledge of the propagation constant inside our fibre. This numerical data will allow us to simplify the calculation of (15) and therefore predict the bandwidth and the mean number of photons per pulse.

3.2. Number of photons per pulse and associated bandwidth

The integral from (15) can be sorted out if we now develop the $\Delta k$-function around the phase-matched frequencies $(2k_{p0} - k_{so} - k_{io} - 2\gamma P = 0)$ using a first-order Taylor power series.

$$\Delta k = 2k_p - k_s - k_i - 2\gamma P$$

$$= 2 \frac{\partial k_p}{\partial \omega} \bigg|_{\omega_p} \Delta \omega_p - \frac{\partial k_s}{\partial \omega} \bigg|_{\omega_s} \Delta \omega_s - \frac{\partial k_i}{\partial \omega} \bigg|_{\omega_i} \Delta \omega_i.$$
Figure 4. The hue colour represents high values of the function. It is revealing to note the angle of the sinc stripe which is controlled by the coefficients $N_i, N_p$—arising from the fibre geometry. This is slightly different from the angle of the exponential stripe which is always fixed at 45°. The difference in angle between the two functions defines the narrowness of the signal and idler bandwidth.

With the help of energy conservation, we can remove the $\Delta \omega_p$ variable and find:

$$
\Delta k = (N_i - N_p) \Delta \omega_{k_i} + (N_i - N_p) \Delta \omega_{k_i},
$$

(20)

where $N_i = [\omega_i \frac{\partial n}{\partial \omega} |_{\omega_i} + n_l]$.

We now have the products of two functions depending only on $\Delta \omega_{k_i}$ and $\Delta \omega_{k_i}$ (similar to spectral functions developed for $\chi^{(2)}$ phase matching [34]): the first one is $\text{sinc}^2(\Delta kL/2)$, standing for the natural phase-matching condition in the PCF for a monochromatic pump. The second one is $e^{-\left(\frac{\Delta \omega_{k_i} + \Delta \omega_{k_i}}{\sigma^2/2}\right)}$ standing for the pump pulse broadening. So if we want to sum over all the possibilities for our fibre, we have to look at the product of these two functions and to integrate it over $\Delta \omega_p$ and $\Delta \omega_i$. We plot in figure 4 the graphical representation of both functions and their product. We can then carry out the formal calculation of $\langle N_i \rangle$ as explained in appendix A to eventually find:

$$
\langle N_i \rangle = \left( \frac{\text{SIL}}{2} \right)^2 \left( \frac{\pi \Delta \omega_p^2}{4 \ln(4)} \right)^{3/2} \left( \frac{4\sqrt{2}\pi c}{(N_i - N_i) L} \right) \frac{1}{v_{g_i} v_{g_i}}.
$$

(21)

If we now develop the $\Delta k$-function around the phase-matched frequencies using first-order Taylor power series but use the energy conservation to remove the $\Delta \omega_{k_i}$ (respectively $\Delta \omega_{k_i}$) term, we get

$$
\Delta \omega_{k_i} = \frac{2\pi c}{|N_i - N_i| L} + 2 \left| \frac{N_i - N_p}{N_i - N_i} \right| \Delta \omega_p,
$$

(22)

$$
\Delta \omega_{k_i} = \frac{2\pi c}{|N_i - N_i| L} + 2 \left| \frac{N_i - N_p}{N_i - N_i} \right| \Delta \omega_p
$$

(23)
giving respectively the bandwidth of the signal and idler photons. It is worth noting two separated terms: the first one corresponding to the natural bandwidth from a monochromatic pump, while the second one is the pump bandwidth broadening. In the classical CW case, even a perfectly monochromatic pump leads to down-converted photon with a finite bandwidth. This process behaves as the inverse of the length of the fibre [35]. Here, we have to take it into account and add the fact that our pump is not monochromatic. The pump bandwidth contribution is linked to the slope (\(\frac{\Delta N_i - \Delta N_p}{\Delta N_s - \Delta N_i}\) for the signal and \(\frac{\Delta N_s - \Delta N_p}{\Delta N_i - \Delta N_s}\) for the idler) of the phase-matching curves in figure 3.

3.3. Experimental limitations

In the previous treatment of the FWM process, we assumed that all the frequency components, and their following quantized operators, were all coherent within the pulse. Our calculation obviously applies to PCF pumped by Fourier transformed limited pulses, nevertheless it would be straightforward to decompose the pulse into several coherent subgroups, apply the treatment to and eventually sum up the number of photons coming from each of them.

Another experimental limitation we did not highlight in the previous treatment is group velocity walk-off between the three involved photons. Although the pump is close to the group velocity dispersion minimum, this is not the case for the signal and idler photons. After a certain distance, the pump pulse no longer overlaps with the pair pulse, thus meaning that we cannot sum up coherently all the operators. Using the propagation constant determined in subsection 3.1, we estimated the walk-off distance to be about 15 cm (for our pulses). Therefore, we have to apply our numerical calculation over this length and eventually to multiply accordingly to cover the actual length of the fibre.

From (22) and (23), we should expect the bandwidths to remain constant beyond this length. Note however, in (21), the number of pairs created always grows linearly with the length. As a consequence, the walk-off distance will not play a role in the number of created photons and monitoring the bandwidth fluctuation versus fibre length is the only way we have to experimentally estimate the figure associated to the walk-off.

4. Experiment

4.1. Setup

In order to estimate the brightness of our source we used the coincidence setup depicted in figure 5 where a mode-locked picosecond Ti : Sapphire pump laser (Spectra Physics—Tsunami) emitting \(~2\) ps pulses with a repetition rate of 80 MHz is sent, through an optical isolator, on to a prism P to remove in-band light from the pump laser spontaneous emission. A pin hole is then used to improve the pump mode and eventually several attenuators bring the power down so that up to 1 mW average power is launched into the fibre. Since the PCF is birefringent and supports two modes, a half wave-plate is used to align the pump polarization along one axis thus preventing polarization scrambling and creating pairs with the same polarization as the pump beam. The output of the fibre is collimated using an aspheric lens, followed by a dichroic mirror centred at 700 nm to spread the incoming beam into two arms, one corresponding to the signal channel and the other to the idler, where band-pass filters F1 and F2 centred at 570 and 880 nm...
Figure 5. Optical layout. Laser, 708 nm Ti:Sa laser; P, prism; HWP, halfwave plate; M, protected silver mirror ($R \leq 95\%$); DM, dichroic mirror (centred 700 nm, $T \leq 85\%, R \leq 90\%$); F1, 570 nm band-pass filter, bandwidth 40 nm, $T = 80\%$; F2, 880 nm band-pass filter, bandwidth 40 nm, $T = 80\%$; APD, silicon single photon detector ($\eta_s \approx 60\%$ and $\eta_i \approx 33\%$). respectively (width 40 nm, $T > 80\%$) are used to remove in-line pump and background light. Each photon of the pair is then launched into single mode fibres that are connected to two silicon avalanche photodiodes (APD). The detected photons are counted in a dual-channel counter and the coincidences between the two APDs are analysed using a time interval analysis system (TIA).

4.2. Results

In order to determine the brightness of our source, we measured the number of single counts in both signal and idler channels, while we recorded the number of coincidences. These coincidences are recorded using a time interval analyser. We present our time interval histograms normalized by the start count rate. This is then the probability that a start count is stopped by a detection in the ‘stop channel’. If the photons are produced always in pairs, then every start detection will have an associated stop photon. The probability of detecting that photon is then the ‘lumped efficiency’ of the optics and detector in the ‘stop channel’. This should remain roughly constant with pump power when we produce less than 0.1 photons per pulse and when background count rates are low. On the other hand, satellite peaks should also arise with the repetition rate of the laser. They should be linked to the probability of having a ‘stop photon’ from a different pair or from background in a neighbouring pulse. The area of the satellite peaks reflects the probability of seeing a stop
Figure 6. Time interval histogram showing the coincident photon detection peak and also a zoom on one of the accidental coincidence peak for different pump powers. The instrument displays the probability that a start pulse is stopped within a given time bin. Here the time-bin width is 156 ps. The time between two peaks reflects the pump laser repetition rate. However, the width of the peaks is limited by the response time of the detectors which is typically hundreds of picoseconds (rather than the actual duration of the pump pulses).

4.2.1. Photon pair rate. The central peak on figure 6 corresponds to signal and idler photons belonging to the same pulse. The small satellite peaks stand for uncorrelated events, i.e. signal and idler coming from subsequent pulses, whether they are actual pairs of photons or background photons. We clearly see in the inset that the satellite peaks grow with pump power, whereas the central peak remains roughly constant. However, in a quantitative analysis we have to take into account both the background and the multiphoton pair rate in the central peak as they increase the apparent photon count probability. Thus we will introduce $C_{\text{raw}}$ as the raw coincidence rate in the central peak and we will calculate the accidental coincidence rate thanks to the satellites
peaks $C_b$. This allows us to write for $C_{\text{raw}}$:

$$C_{\text{raw}} = \mu_s \eta_s \mu_i \eta_i r + C_b,$$

where $r$ is the actual photon pair rate; $\eta_s$ and $\eta_i$ are the APD quantum efficiencies at 586 and 894 nm, while the net optical transmission and launch efficiencies into single mode fibre of each arm are $\mu_s$ and $\mu_i$. We defined the single counting rates in the signal and idler APDs as:

$$N_{\text{raw}}^s = \mu_s \eta_s r + \sum_{n=2}^{\infty} \alpha_n \mu_s \eta_i r^n + B_s,$$

$$N_{\text{raw}}^i = \mu_i \eta_i r + \sum_{n=2}^{\infty} \alpha_n \mu_i \eta_i r^n + B_i,$$

where $B_s$, $B_i$ are total background rates and $\alpha_n \mu_s \eta_i r^n$, $\alpha_n \mu_i \eta_i r^n$ stand for the multiphoton pair contributions ($\alpha_n$ represents the increased probability of detecting any one of $n$ photons). Here, we separated the background and multiphoton contribution in (25) and (26), as we can already consider the latter being negligible for low average powers. For instance, as long as we remain under 0.1 photon pairs per pulse, the high order contribution should remain a tenth of the total. On the other hand, the background might not be negligible and is mostly due to Raman scattering in our case [29]. Assuming we can single out this background contribution, as shown in [36], we can now use the single counting rates and the coincidence rates to estimate the actual rate of pairs $r$ created inside the PCF using this equation:

$$r = \frac{(N_{\text{raw}}^s - B_s)(N_{\text{raw}}^i - B_i)}{(C_{\text{raw}} - C_b)}.$$

As previously stated, $B_s$ and $B_i$ include the Raman background rates in the APDs and we have to single out these detections. The number of created photon pairs is proportional to the square of the peak intensity and the Raman scattering grows roughly linearly at these relatively low pump powers. Thus switching the laser from the pulsed regime to the CW regime reduces the photon pair rate to negligible levels while keeping the Raman and other sources of background constant for the same average power. We propose to validate the technique by recording the number of events in the two APDs versus the pump power for each case (i.e. pulsed and CW) and studying the evolution of the counting rate summarized in table 1. Here a linear tendency would be the proof of spontaneous Raman scattering, while pure quadratic behaviour would be the proof of actual photon pairs. All the plotted data have been corrected to compensate the nonlinearity of the Si-APD for high counting rate following the datasheet [37].

We first plot in figure 7 the number of detections versus the pump power in CW regime. We clearly see the negligible level of Raman scattering at the signal wavelength compared to the idler wavelength, as expected as the anti-stokes Raman scattering is often considered negligible compared to the stokes one. In both cases, the fit is purely linear (quadratic term is negligible) and being the proof of spontaneous Raman scattering process and possible leakage of pump light. We can therefore use these figures as a good estimation of the Raman background rate for any given mean pump power.
Table 1. Experimental measurement of the coincidence rate versus the pump power for 0.2 m of PCF. The dark count rate in both APD is $\sim 400$ Hz. Note that the central peak percentage ($C_i/N_i^{\text{raw}}$) remains almost constant. The small increase corresponds to the increasing percentage of the satellite peaks ($C_b/N_i^{\text{raw}}$) that affect little the central peak probability.

| Power ($\mu$W) | $N_i^{\text{raw}}$ (kHz) | $N_i^{\text{CW}}$ (kHz) | $N_b^{\text{raw}}$ (kHz) | $N_b^{\text{CW}}$ (kHz) | $C_i$ (kHz) | $C_b$ (kHz) |
|---------------|--------------------------|--------------------------|--------------------------|--------------------------|-------------|-------------|
| 960           | 2070                     | [5.0]                    | 917                      | [54]                    | 234         | [16]        |
| 660           | 1031                     | [3.0]                    | 460                      | [38]                    | 115         | [3.8]       |
| 490           | 601                      | [2.0]                    | 261                      | [25]                    | 66          | [1.4]       |
| 340           | 298                      | [1.2]                    | 135                      | [18]                    | 33          | [0.1]       |
| 200           | 107                      | [0.9]                    | 52                       | [11]                    | 12          | [0]         |
| 0             | 0.4                      | [0.4]                    | 0.4                      | [0.4]                   | 0           | [0]         |

Figure 7. Corrected raw counting rate as function of the pump power in both APDs in CW regime for 0.2 m of fibre.

We then plot in figure 8 the number of net detections versus the pump power. We define the net detections as being the number of detections in pulsed regime minus the number of detections for the same average power in CW regime. We clearly see that the number of net detection versus the pump power is purely quadratic, thus highlighting that we are dealing with actual photon pairs. We can then now identify $B_s$ and $B_i$ as the CW counting rate and calculate in table 2 the actual number of photon generated in the fibre using (27). Note that the raw counting rates can be used, once we have $B_s$ and $B_i$ this is an absolute method for determining the photon pair rate. If we eventually compare the counting rate associated with the actual Raman scattering to the counting rate associated with the actual photon pair, it confirms that the background in the signal channel is extremely low ($\leq 0.3\%$ of the total rate) while in the infrared we estimate the background rate to remain relatively low ($\leq 6\%$).

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Figure 8. Corrected net counting rate as function of the pump power in both APDs in pulsed regime for 0.2 m of fibre. The net counting rate stands for the raw counting rate minus the CW counting rate. We added here the CW counting rate as a guide for comparison with figure 7.

Table 2. Experimental estimation of the photon pair rate versus the pump power for 0.2 m of PCF. From left to right, we first recall the associated pump power and the theoretical photon pair rate using (21), followed by the experimental photon pair rate using (27). For information purpose, we added the mean number of photon per pulse \((r_{\text{exp}})/(R_{\text{laser}})\), the coincidence to accidental contrast \((C_{\text{raw}} - C_{b})/(C_{b})\) and the lumped probability of detecting respectively the signal \((\mu_s \eta_s)\) and idler photons \((\mu_i \eta_i)\).

| Power (µW) | \(r_b\) (s⁻¹) | \(r_{\text{exp}}\) (s⁻¹) | \(\langle n_{\text{exp}}\rangle\ | C : A | \(\mu_s \eta_s\) | \(\mu_i \eta_i\) |
|-----------|----------------|----------------|----------------|--------------|-------------|-------------|
| 960       | \(\sim 8.05 \times 10^6\) | 8.46 \times 10^6 | 0.11 | 15:1 | 0.235 | 0.106 |
| 660       | \(\sim 3.81 \times 10^6\) | 4.08 \times 10^6 | 0.05 | 36:1 | 0.240 | 0.109 |
| 490       | \(\sim 2.10 \times 10^6\) | 2.31 \times 10^6 | 0.03 | 55:1 | 0.244 | 0.109 |
| 340       | \(\sim 1.10 \times 10^6\) | 1.14 \times 10^6 | 0.015 | 220:1 | 0.244 | 0.109 |
| 200       | \(\sim 0.35 \times 10^6\) | 0.43 \times 10^6 | 0.006 | – | 0.223 | 0.109 |

Considering table 2, if we compare the photon pair rate to the theoretical rate, we have here good agreement. However, one has to bear in mind that the estimation of the mode area and the peak power could lead to large error in the expected pair rate \(r_b\). We found it useful to recall the lumped probability to detect the photons of the pairs. These important experimental parameters are useful for evaluating the possibility to perform multiphoton pairs experiment as discussed in section 5. Here, provided a detection efficiency \(\eta_s \approx 0.60\) and \(\eta_i \approx 0.33\) [37], we found a global coupling efficiency from the PCF fibre to the single mode fibre of \(\mu_s \approx 0.42\) and \(\mu_i \approx 0.30\). These figures include the fibre coupling, filters, dichroic mirror and lenses losses for signal and idler channels respectively. By correcting these results with the filters and dichroic mirror losses,
we find a coupling efficiency into single mode fibre of respectively 0.58 and 0.44 for the signal and idler photons. We also present in table 2 the coincidence to accidental contrast \((C:A)\) which has in the past been used [38] to describe the quality of any presented coincidence rate. In our experiment the background rate is quite low and our \((C:A)\) reflects the random overlap of pairs of pairs. We discuss this further in section 5.

4.2.2. Walk-off. Taking into account the walk-off distance we identified in subsection 3.3, we should expect the bandwidth to be constant versus the fibre length whenever the fibre is longer than 15 cm. For several fibre lengths, we recorded the pump bandwidth, measured the signal fluorescence bandwidth and compared it to (22). From figure 9, one can note the quite good matching between the measured and predicted bandwidth and the similar behaviour of both bandwidths versus the fibre length. As predicted, the measured bandwidth remains constant whenever the fibre is longer than the walk-off distance which we estimate to be around 15 cm. More measurements around 15 cm would be needed to reduce the uncertainty about the walk-off experimental determination.

5. Discussion

This source has to be usable for future quantum interference experiments involving three or more photons created as two or more pairs [30]. We dedicated the following section to estimate the potential of this source. To do so, we used realistic requirements for interference effects between separate pair-photons and attached experimental figures to a typical four-photon quantum experiment.
5.1. Multiphoton pair weight in satellites peaks in figure 6

We showed in the previous sections that the spontaneous Raman scattering was negligible compared to the photon pair rate. However, in a multiphoton experiment, the probability of creating two photon pairs has to dominate the probability of creating one photon pair and a Raman photon. We can use figure 10 to state that our source exhibits a background low enough to allow multiphoton pairs experiments. We plotted the background coincidence rate $C_b$ (table 1) versus the pump power. The spontaneous Raman scattering rate grows linearly with pump power whereas the photon pair rate grows quadratically. Thus the background coincidence rate (associated to the satellites peaks) should reflect the weight of each contribution. We fitted the experimental data with a polynomial function and it clearly appears that the satellite counting rate is dominated by the multiphoton contribution ($\propto P_p^4$). It means these peaks are thus indicative of the four-photon coincidence rate we will get in future experiments and are variables we want to maximize.

For these multiphoton quantum information experiments, we require a narrower bandwidth so that the coherence length is equivalent to the pulse length [30]. A quantum interference experiment involving fourfold coincidence between photons coming from two separated sources would require a filter of order 0.2 nm bandwidth in the green (0.4 nm in the IR). Such filters will transmit only 40% of in-band light thus halving our effective efficiencies and collect only $\sim 1/25$ of the available spectrum. However, our present coincidence rates are limited by the detector saturation. Here, the single counting rates would be significantly reduced thus allowing an increase in pump power. Using our source with a pump power of $\sim 6$ mW, the expected rate of photon pairs detected within this bandwidth is $\sim 9 \times 10^4$ s$^{-1}$, which means a rate of four photon events $\sim 100$ s$^{-1}$, two orders of magnitude higher than any previous experiment. It is also important to underline, that our Raman background rate in the APDs, will be reduced by a bandwidth factor of $\sim 1/100$ due to its broadband nature thus improving even more the performances of the source.
6. Conclusion

We have presented in this paper both theoretical and experimental aspects of photon pair generation using FWM in a PCF. We have clearly demonstrated a good agreement between the theory and experiment. Any realistic multi-photon quantum experiment requires a precise timing using ultra-short pulses, thus we have developed a quantum model suitable for such a regime. We used it to suggest a numerical estimation of the photon pair rate from a PCF provided a good knowledge of the mode propagating in the fibre. We have validated the model by comparison with the experimental measurement of picosecond-pulsed photon pairs generated by FWM in a single-mode optical fibre. The source we used is polarized, bright, narrowband, single-mode and tuneable by varying laser wavelength or fibre parameters. The wide separation of the generated pair wavelengths means that most of the background can be avoided. All these advantages make this new source of photon pairs extremely promising for quantum information processing applications.

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Appendix A. Estimation of the number of photons

From (15)

\[
\langle N_s \rangle = \left( \frac{SIL}{2} \right)^2 \left( \frac{\sqrt{\pi}}{\sigma} \right)^2 \int \int \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) e^{-\frac{(\Delta \omega_{s1} + \Delta \omega_{s2})^2}{2 \Delta \omega_{s}^2}} \frac{d \Delta \omega_{k_s}}{v_{g_s}} \frac{d \Delta \omega_{k_i}}{v_{g_i}}.
\]

This integral can be numerically evaluated but an approximate analytic form can be obtained if we develop the \( \Delta k \)-function around the phase-matched frequencies using first-order Taylor power series:

\[
\Delta k = -N_p (d \omega_{p1} + d \omega_{p2}) + N_i d \omega_i + N_s d \omega_s,
\]

where \( N_i = [\omega_i \frac{d n_i}{d \omega}]_n + n_i \). Then with the help of energy conservation \((-d \omega_{p1} - d \omega_{p2} + d \omega_i + d \omega_s = 0\) we find:

\[
\Delta k = \frac{(N_s - N_p)}{c} d \omega_s + \frac{(N_i - N_p)}{c} d \omega_i.
\]

We can now write equation (15):

\[
\langle N_s \rangle = \left( \frac{SIL}{2} \right)^2 \left( \frac{\sqrt{\pi}}{\sigma} \right)^2 \times \int \int \text{sinc}^2 \left( \frac{(N_s - N_p) L}{2c} \Delta \omega_{k_s} + \frac{(N_i - N_p) L}{2c} \Delta \omega_{k_i} \right) \times e^{-\frac{(\Delta \omega_{s1} + \Delta \omega_{s2})^2}{2 \Delta \omega_{s}^2}} \frac{d \omega_{s}}{v_{g_s}} \frac{d \omega_{i}}{v_{g_i}}.
\]
It is quite easy to integrate this product making the following change of variables ($X = \Delta\omega_k + \Delta\omega_k / 2$ and $Y = \Delta\omega_k - \Delta\omega_k / 2$). Then it becomes obvious that if we perform the $Y$ integral first and consider the sinc function as a gate of height 1 and FWHM $= ((N_f - N_i) L / (2c)) \Delta Y = \pi$, the system is then easy to solve and we find:

$$\langle N_s \rangle = \left( \frac{SIL}{2} \right)^2 \left( \frac{\sqrt{\pi}}{\sigma} \right)^2 \left( \frac{2\pi c}{(N_f - N_i) L} \right) \frac{4}{v_g v_g}, \quad (A.5)$$

$$\langle N_s \rangle = \left( \frac{SIL}{2} \right)^2 \left( \frac{\pi \Delta\omega_p^2}{4 \ln(4)} \right)^{3/2} \left( \frac{4\sqrt{2}\pi c}{(N_f - N_i) L} \right) \frac{1}{v_g v_g}, \quad (A.6)$$

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