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# Continuous-Wave generation of photon pairs in silica nanofibers using single- and multi-longitudinal mode pumps

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## **Abstract :**

In this paper, we describe experimental observation and characterization of emission of photons pairs through FWM in a silica nanofiber, with high Coincidence to Accidental Ratio (at best greater than 20000 in the CW regime at pump power 24mW and 40000 in the pulsed regime at pump around 150 $\mu$ W) and high pair emission rate (above 1MHz in the pulsed regime with CAR around 100 at a pump power around 7mW), despite residual presence of Raman spontaneous scattering (in the vicinity of the 12<sup>th</sup> silica Raman band) on the Idler side. Moreover, these pairs of photons are observed with either single longitudinal mode source or a multimode source (as well as a pulsed source) showing a factor of two increase of emission rate with the multimode source thanks to its random temporal fluctuation coupled to the quadratic response of FWM.

## I Introduction

Source of photon pairs are traditionally based on Spontaneous Parametric Down-Conversion which can be indifferently operated either in Continuous Wave (CW) or in the pulsed regime [1, 2]. Synchronous and Asynchronous Quantum protocol were then developed in both regimes, such as the ones using Energy-Time entanglement in the CW regime and its equivalent in the Pulsed regime, the Time-Bin entanglement [3, 4]. With the development of Spontaneous Four-Wave Mixing integrated sources, both regimes continue to be explored depending on the used structures, pulsed operation in waveguides [5], and CW operation in resonant systems (Ring cavity, CROW waveguides, ...) [6]. For fibered structures, the easier to operate pulsed regime stays the preferred regime used in applications and the Continuous Wave (CW) regime fibered generation of pair of photons stays a long search that until now only gave few results [7, 8, 9, 10, 11].

There are several reasons for that difficult search. The main one is related to the quadratic power response of the four-wave-mixing (FWM) mechanism used to generate these photon pairs that leads to much less efficiency and lower generation rates compared to the preferred pulsed regime. The second reason is the simultaneous presence of Spontaneous Raman Scattering, that generates a large quantity of noise photons that will mask the pairs. The broadband nature of the Raman emission spectra of silica, compared to crystalline materials such as Silicon [5, 6, 12], to liquids [13], or even to materials such as noble gases that do not present Raman transitions [14, 15], will prevent any frequency filtering of these photons. This will be thus a major limitation of the use of silica fibers as sources of pairs of photons. Due to its linear power dependence, spontaneous Raman scattering is all the more a problem in the CW regime that often prevents to see the much smaller quadratic signature of the number of detected photons in CW experiments [9] (even if the use of the coincidence detection reveals the pairs). In order to eliminate the deleterious influence of Raman scattering in silica fibers, low temperature operation [8, 9, 11] can be used, with the drawbacks of a higher experimental complexity. Another possibility is the use of a large frequency shift of the photon pairs [7, 10] but with incomplete elimination due to the residual presence of higher order Raman bands [7].

Large frequency shifts are generally obtained by using either photonic crystal fibers [7, 10, 16], that thanks to their small core diameters present properly positioned Zero Dispersion

Wavelengths, or birefringent fibers [17] using vectorial four-wave mixing. One possible alternative is the use of tapered micro and nanofibers [18, 19, 20]. The nanofibers are easily fabricated from a standard fiber by pulling it above a flame, the nanofiber waist size and length, can be chosen any value in the micrometer and the sub micrometer range [21], with length that can reach several centimeters (until some tens of centimeters [22]). The nanofiber is conceived and fabricated together with the tapers that connect it to the standard fibers, which means that extremely high transmission (typically greater than 90%) can be obtained and potentially lossless coupling of photons in and out of the nanofiber can be obtained. Pulsed generation of photon pairs in micro-nanofibers had already been demonstrated and is still the subject of intense research [23, 24, 25, 26, 27], nevertheless generation of photon pairs in the CW regime has to the best of our knowledge not been observed in these nanofibers until the results presented in this paper.

Moreover, we demonstrate that the pair emission rate in the CW regime depends on the temporal modal contents of the pump, with a two times higher emission rate measured with a multimode pump. The experiment is based on two classical laser sources used in nonlinear optic experiments, first a tunable pulsed (picosecond) mode-locked Ti-Sapphire Laser, and a Distributed Bragg Reflector (DBR) single-longitudinal mode Laser diode. Both sources emit in the near infrared and are used around 852nm (the laser diode wavelength). The Ti-Sapphire laser (Tsunami from Spectra-Physics) is pumped by a CW green laser and is actively mode-locked to deliver short pulse with pulse duration in the picosecond range (autocorrelation FWHM duration around 2 to 6ps) at a high repetition rate of 80MHz. When mode-locking is disabled, the emitted beam becomes CW (with the same average power), but the laser emits light in a highly multi-longitudinal mode regime (necessary to obtain short pulses by locking all the mode), and the beam presents high speed random temporal fluctuations that average to the mean power. We will thus be able to directly compare our single photon source operation in these three temporal regimes (mode locked pulses, multi-longitudinal mode CW and single-longitudinal mode CW).

## II Choice of the nanofiber characteristics

This pump is sent in the nanofiber that will be our nonlinear medium. Photons Signal (S) and Idler (I) frequencies are created with a spectral probability of emission characterized

by the Joint Spectral Intensity (JSI) [28] that writes as the product of two functions related to energy ( $2\omega_p - \omega_s - \omega_I = 0$ ) and momentum ( $2\beta_p - \beta_s - \beta_I = 0$ ) conservation (Appendix A). The size and length of the nanofiber are chosen to have best performances with the chosen pump wavelength (around 852nm) in order to generate an idler photon in the telecom band (around 1550nm), and thus a signal photon being through energy conservation in the visible range (around 580-590nm). The simulation (Appendix B) shows that for our experimental conditions the nanofiber radius has to be in the 412nm range [23]. The length of the nanofiber should be as long as possible to reach a high emission efficiency, but with the constraint that the nanofiber radius should stay uniform within some tenth of percent on that range. In our case, a nanofiber length of 1cm appeared to us a good compromise considering the performance of our pulling platform to realize a good generation rate (at the expense of a slightly higher pump power to compensate for the reduced length) and a near ideal phase matching Sinc shape.

The nanofiber is connected to the initial SMF28 fiber by tapers that are realized simultaneously with the nanofiber during the "Pull and Brush" fabrication procedure. The procedure ability to effectively realize the designed nanofiber and tapers had been previously validated, using a home-made shape measurement optical system [21] and scanning electron microscope measurements [29]. We designed the tapers with parameters giving symmetric adiabatic tapers (21 mm length each) that allow to realize a virtually lossless coupling of light from the fundamental mode of the SMF28 fiber core to the nanofiber (for more details about conditions to be fulfilled in order to obtain adiabatic tapers the reader can see for example ref. [30]). The nonlinear object thus consists in connectorized SMF28 access fiber (typical length 40cm), the entrance taper followed by the nanofiber (radius 412nm, length 1cm) and the output taper and a second connectorized SMF28 access fiber (typical length 40cm) (see insert in Fig. 1). The ensemble (tapers and nanofiber, with a global length of 5.2cm) is fixed in a glass box to manipulate it easily and to protect it, as best as possible, from ambient dust.

Once a nanofiber is pulled and prepared with fiber connectors, we first realize a transmission measurement test, to verify the nanofiber integrity and have a first idea of its quality and of our ability to inject the pump preferentially in the fundamental HE11 mode of the nanofiber (even if the nanofiber and the SMF28 are slightly multimode, with a V parameter at pump wavelength around 4 in both cases, careful injection of light allows to excite only the fundamental mode, with negligible coupling to high order modes during propagation thanks

to the adiabatic tapers). The typical raw transmission of a nanofiber (corrected from optical element losses) on our experimental set-up is around 50 - 60%. It is, to habitual experimental alignment fluctuations, identical to the one obtained with an unpulled SMF28 fiber prepared identically. This transmission is essentially limited by the Fresnel reflection losses of fiber connectors, by the quality of fiber cleaving, by the residual aberration present on the laser pump beam and by the alignment quality. Freshly realized nanofibers have thus virtually no propagation and tapers coupling losses. Nevertheless, after some time (i.e. at least after several months), we observe some decrease of the nanofiber raw transmission, probably due to accumulation of dust in the vicinity of the nanofiber waist (correlated to apparition of punctual scattering points on the nanofiber waist, not initially present on the freshly pulled fiber). After almost one year the raw transmission of the nanofiber used in this study decrease around 30%, attributed to propagation losses in the nanofiber waist part with a transmission of the nanofiber going from near 1 to around 0.6, indicating that the protecting glass box was not totally hermetic [31].

### III Experimental set-up

The nanofiber is inserted in the experimental set-up shown in Fig. 1. The pump beams are coupled to the access entrance fiber together with probe beams at 633nm (He-Ne laser) and at telecom wavelengths (1520nm – 1620nm tunable laser diode) using a dichroic mirror. These additional lasers are used for alignment optimization of the detection channels. The tunable laser diode is additionally used to characterize the photon pair emission spectrum (Joint Spectral Intensity) through a Stimulated Emission Tomography (SET) experiment [32, 33, 34]. The pump beam power is varied before the injection using a  $\lambda/2$  – Polarizer set-up. At the output of the nanofiber (i.e. at the output of access SMF28 fiber) the beam is collimated and send to dichroic mirrors first to extract the high-power pump beam, and then to separate the signal and idler photons. Additional high and low pass filters as well as interferential band pass filters are used to further eliminate residual scattered pump photons. Signal photons are coupled to a SM780 fiber, whereas Idler photons are coupled to a SMF28 fiber and coupled to additional fibered filters (CWDM filters or tunable filter). Then the fibers are connected to single photon detectors, a Geiger mode silicon Avalanche photodiode (APD) for the signal photon detection (Aurea technology module) and a Geiger mode InGaAs APD (ID230 module

from IDQuantique) for idler photon detection. Both APDs deliver TTL pulses corresponding to the detected photons and are connected to a computer-controlled counter and temporal correlator devices (ID900 module from ID Quantique). Fibers can also be connected to power-meters or Optical spectrum analyzers and fibered spectrometer, for set-up characterization and SET measurements. Extracted pump beam is also sent to a multimode fiber for pump spectrum measurement or to a power-meter for transmitted power measurement.

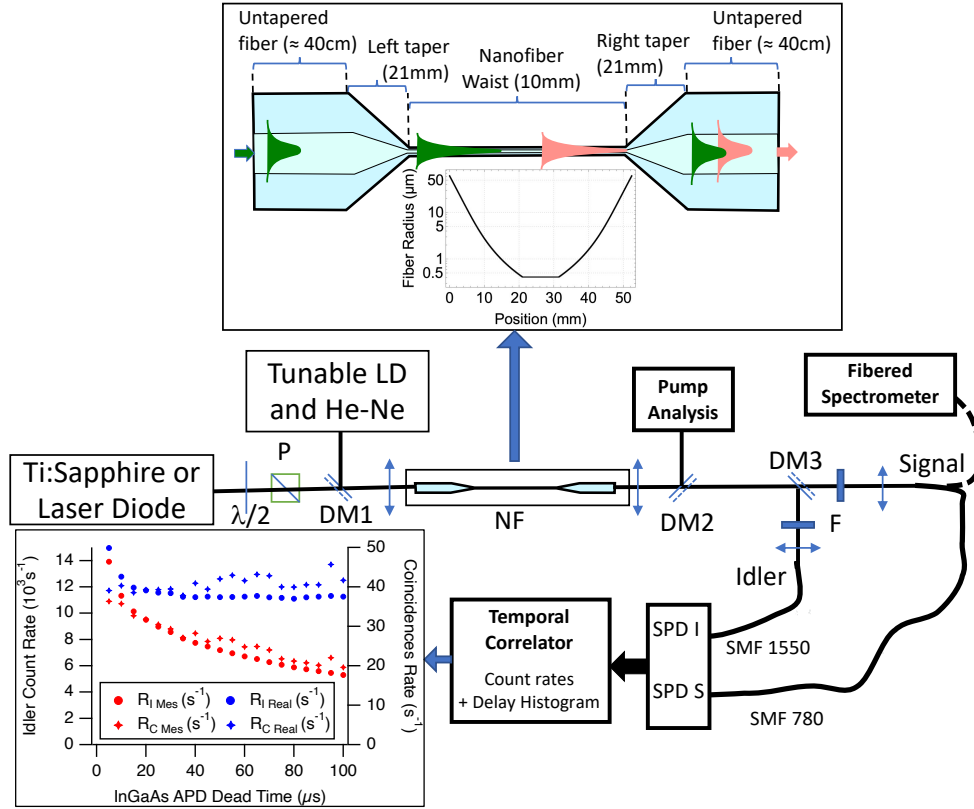


Figure 1: Schematic of the experimental set-up (DM: Dichroic Mirrors, P: Polarizer,  $\lambda/2$ : Half wave plate, F: High Pass, Low Pass or Band Pass spectral Filters). Light collected from the nanofibers is injected in single mode Fibers (SMF), connected to different characterization instruments, and to Single Photon Detectors (SPD) for Idler and Signal wavelengths, followed by a temporal correlator for counts measurement and coincidence analysis (additional fibered components, CWDM or tunable filters, couplers, not shown here, might also be used). The upper insert shows the global nanofiber structure, with a plot of the designed shape of the adiabatic tapers realized by the pulling platform. The lower insert shows a plot of real (Blue/Dark Gray symbols) and measured (Red/Gray symbols) detector count and coincidences rates showing the effect of deadtime correction on the acquired data.

We thus measure the signal and Idler photons count rates,  $R_{S\ Mes}$  and  $R_{I\ Mes}$  (in  $s^{-1}$ ) respectively, received by the two detectors, as well as coincidence histograms between the signals of the two detectors from which we deduce the coincidences rate  $R_{C\ Mes}$  (in  $s^{-1}$ ) from the number of samples detected in the coincidence peak (on a temporal window  $t_{bin} = 1.3ns$  corresponding to the width of the coincidence peak) and normalized to the measurement duration (typically between 10s and 1 hour depending on measurement power and temporal

regime). These measured data are corrected for the deadtime of the detectors ( $\tau_{DT I} = 20\mu s$  and  $\tau_{DT S} = 33ns$ ) to give the actual count and coincidences rates:

$$R_{S,I Real} = \frac{R_{S,I Mes}}{(1 - \tau_{DT S,I} R_{S,I Mes})} \quad (1)$$

$$R_C Real = \frac{R_C Mes}{(1 - \tau_{DT S} R_S Mes)(1 - \tau_{DT I} R_I Mes)} \quad (2)$$

that will be the ones analyzed in final. We have verified these expressions (see lower insert in Fig. 1), at least for the InGaAs detector, which deadtime can be adjusted and for which deadtime correction is more critical. The real value calculated for these experimental conditions are then deadtime independent (contrarily to the measured value) except for the detector count rate at low deadtime where the influence of the detector after-pulses become dominant (all the presented data in the paper used  $\tau_{DT I} = 20\mu s$  chosen as a compromise to prevent detector saturation at high counting rates and a moderate after-pulse probability value around 10% [35] that allows to neglect their influence in first approximation).

#### IV Photon pairs emission in CW and pulsed regime

The nanofiber is first characterized using a Stimulated Emission Tomography (SET) [32] experiments (Appendix C), for an easy and rapid measurement of the Joint Spectral Intensity (JSI) that characterizes the emission spectra of the source. This allows a precise alignment of the experiment. The measured JSI is the same whatever the pump temporal regime and is very close to the expected Sinc shape, with nevertheless small side lobes attributed to fluctuation of the nanofiber diameter [36]. That shape is compatible with the one caused by a sinusoidal fluctuation (Appendix D) of the Zero Dispersion Wavelength (ZDW) with a period of 3.83mm and an amplitude of 2.2nm (around the reference value of  $\lambda_{ZDW} = 1036nm$ ). Such a variation of the ZDW corresponds to a variation of the diameter of the nanofiber of about  $\pm 2$  nm, around the mean diameter of 824nm, showing a very good uniformity of the nanofiber.

Then, the signal and Idler photons count rates,  $R_S$  and  $R_I$ , as well as coincidence histograms between the signals of the two detectors from which we deduce the coincidences rate  $R_C$  (in  $s^{-1}$ ) are measured as a function of the mean power of the pump beam for the different temporal regimes. The results are shown in Fig.2.



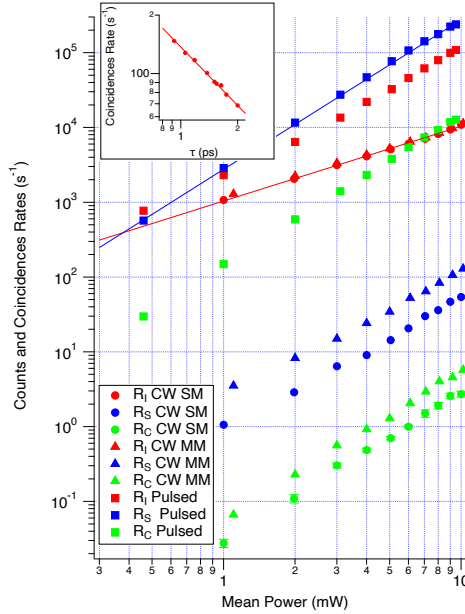


Figure 2: Intensity dependence of the Idler count rate (red/Gray symbols), Signal count rate (Blue/Dark Gray symbols) and coincidences rate (Green/Light Gray symbol), for the Pulsed pump source (Square symbols), the longitudinally multimode CW pump source (CW MM, Triangle symbols) and the longitudinally Single mode CW pump source (CW SM, Disk symbols). The lines show the main quadratic (blue/dark gray line) and linear (red/gray line) power dependences. The mean power is measured at the nanofiber output, after the dichroic mirror used to extract the pump beam. The presented Idler and Signal count rates are subtracted from the independently measured dark count rate of the used detectors ( $\text{DCR}_I = 65\text{s}^{-1}$  and  $\text{DCR}_S = 53\text{s}^{-1}$ ). The errors bars on the measurement are about the size of the symbols and are only shown for the coincidences rate of the single mode CW pump. The autocorrelation FWHM of the pulse for this experiment was 2.8ps (corresponding to a pulse duration  $\tau = 1.03\text{ps}$  for the hyperbolic secant pulse  $I(t) = I_0 \text{sech}^2\left(\frac{t}{\tau}\right)$ ). The insert shows the variation of the coincidences rate as a function of the pulse duration with an adjustment with a function  $A \cdot \tau^{-1}$ .

#### IV-A Analysis of photon pairs measurement

Firstly, we see on Fig. 2 a clear quadratic response (line of slope 2 on the used log-log scale) for the different quantities except for the Idler detector in the CW regimes (red/gray triangle and disks), that presents a linear response. Moreover, that idler count rate is almost identical for both CW pump source (either single or multimode). This indicates that despite the very high frequency shift (almost 700nm between idler and pump) the Idler counts are still dominated by the Raman response of the silica nanofibers, even if for that frequency shift the idler is in the vicinity of the 12<sup>th</sup> silica Raman band (i.e. much farther than in previous measurements [7, 23]). In the pulsed regime, as expected, the FWM quadratic contribution is much higher and clearly dominates. On the contrary, on the signal side (blue/dark gray triangle and disks) the quadratic dependence indicates a complete absence of Raman scattering contribution (which is, with such shifts, naturally orders of magnitude smaller on the Anti-Stokes side than on the Stokes side) and the quadratic response is observed even in the CW regime at very low power (after correction from the detector dark count contribution). This is

the first time that the quadratic response of detector count rate is seen in the CW regime for fibered structure, as the Raman contribution on the anti-Stokes side prevented to reach the quadratic behavior in previous results from the literature [9].

In the pulsed regime the clear quadratic evolution of counts and coincidence shows that we can consider we are in an almost Raman free FWM contribution similar to the one studied in [15]. By comparing coincidences rate ( $R_C = \eta_S \eta_I \mu_P \bar{I}^2$ , with  $\bar{I}$  the mean pump power, and  $\mu_P$  the pair generation efficiency coefficient at the nanofiber exit) to Signal and Idler count rates ( $R_{S,I} = \eta_{S,I} \mu_P \bar{I}^2$ ) in this pulsed regime, we deduce the signal ( $\eta_S = 12\%$ ) and idler ( $\eta_I = 5\%$ ) detection efficiencies of our measurement set-up (often identified to the heralding efficiency). These detection efficiencies include the quantum efficiency of the detectors ( $QE_S = 56\%$  and  $QE_I = 21\%$ ) and the transmission losses of the detection channels (that consisted in the optical losses of the different optical elements for about half of the losses, and the injection losses of the detected beams in the collection single mode fibers of our fibered single photon detectors, injection losses that are typically lower than 50% depending on alignment optimization). All the measurement of Fig. 2 having been realized on the same day without requiring any readjustment of the detection stage (only the pump injection in the nanofiber had been possibly reoptimized when switching between different temporal regimes) these values are then supposed to be identical in the CW regime (the previous simple treatment is not possible in this regime due to the Raman contribution).

To go beyond this first rapid and qualitative analysis of the response in the different regimes, we adjust all the experimental data presented in Fig. 2 with a 2<sup>nd</sup> order polynomial function. We here concentrate on the most significant results for our quantitative analysis (the complete analysis and results of these adjustments are presented in Appendix E). The linear coefficient term of the Idler count rate will be related to the Raman contribution, we found values  $1031 \pm 14 \text{ s}^{-1} \text{mW}^{-1}$  with the CW Single mode source,  $1088 \pm 14 \text{ s}^{-1} \text{mW}^{-1}$  with the CW multimode source and  $1183 \pm 41 \text{ s}^{-1} \text{mW}^{-1}$  in the pulsed regime. This coefficient is found very close in the three regimes as expected for an effect that is directly related to the mean power of the pump beam. The second important parameter is the quadratic coefficient term of the coincidences rate related to the four-wave mixing nonlinear efficiency. We found values  $0.0291 \pm 0.0028 \text{ s}^{-1} \text{mW}^{-2}$  with the CW Single mode source,  $0.0570 \pm 0.0044 \text{ s}^{-1} \text{mW}^{-2}$  (i.e. 1.96 times higher) with the CW multimode source and  $146.5 \pm 3.4 \text{ s}^{-1} \text{mW}^{-2}$  (i.e 5034 times higher) in the pulsed regime. As expected, the pair generation efficiency is higher in the pulsed regime,

but we also measure a two times higher efficiency with the multimode source compared to the single mode source. Considering the detection efficiencies, the generated pair rate at the output of the nanofiber reaches  $\mu_p = 24 \times 10^3 \text{ pairs.s}^{-1}\text{mW}^{-2}$  in the pulsed regime, i.e. at 10mW power a pair generation rate of 2MHz (Fig. 3) that corresponds to less than  $2.5 \times 10^{-2}$  pairs per pulse, a value sufficiently low to consider the generation of multiple pairs as negligible. This rate is similar to the one already observed in nanofibers [24] with just a slightly higher operating power allowing to compensate for the smaller nanofiber length and the longer pulse duration. In the CW regime we have, as expected for a nonlinear mechanism with quadratic response, a lower generation rate  $\mu_p = 5 - 10 \text{ pairs.s}^{-1}\text{mW}^{-2}$  which would correspond to a generation rate of the order of 12kHz at a power of 35mW, a value which is only 25 times smaller than the one of a commercial SPDC source [37] (with a possibility to close the gap using a higher pump power in the range of 200mW than can be accessible relatively easily with usual sources, especially if we consider that multimode pumps are beneficial in our case).

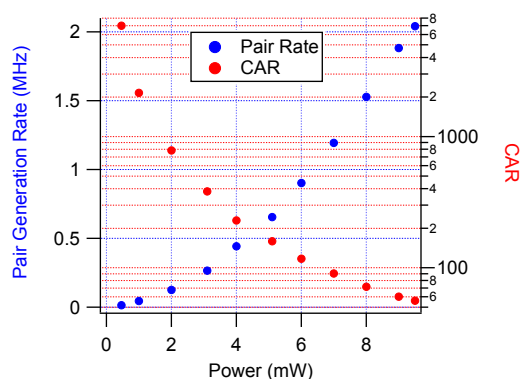


Figure 3: Plot of the pair generation rate (i.e. coincidences rate corrected from detection efficiencies), and CAR in the pulsed regime pumped at 852nm. The generation rate reaches 2MHz with a CAR value of 56, at a moderate pump power of 9.5mW. This generation rate is similar to the one obtained in the literature [24], with at least a three times higher CAR, the higher power used here (9.5mW, instead of 2mW) allows to compensate for the smaller length of the nanofiber and the slightly longer pulse duration.

#### IV-B Source intensity-distribution influence on pair generation

The change of nonlinear performances with the modal content of CW sources had already been observed and analyzed in the context of classical nonlinear optics [38, 39]. It is observed here for the first time to our knowledge in the context of quantum source of pair of photons based on a nonlinear phenomenon. This change of behavior is due to the fact that the temporal statistic of the highly multimode source is close to the one of a thermal source

[40, 41] and that the nonlinear response will be affected by that statistics. The multimode source presents temporal spikes that may have high peak powers and that will generate nonlinearly more pairs on short period of time. To quantify this effect and estimate the efficiency in the different regimes, we use the model of reference [40] in which it is shown that the nonlinear efficiency of a nonlinear mechanisms which depends on the  $n^{\text{th}}$  power of a pump beam intensity is related to the  $n$  order moment ( $\langle I^n \rangle = \int_{-\infty}^{\infty} I^n P(I) dI$ ) of the intensity distribution  $P(I)$  describing the pump beam statistics (and verifying  $\int_{-\infty}^{\infty} P(I) dI = 1$ ). For the single mode CW pump, we have evidently  $P(I) = \delta(I - \bar{I})$ , and for the multimode source, we use the usual expression  $P(I) = \frac{1}{\bar{I}} e^{-\frac{I}{\bar{I}}}$  of a thermal source [38, 39, 40]. Finally, for the pulsed source we rederived the expression  $P(I) = -\frac{2}{T} \frac{1}{dI/dt}$  developed for gaussian pulsed source [40] to the case of our pulsed source that delivers hyperbolic secant pulses  $I(t) = I_0 \operatorname{sech}^2\left(\frac{t}{\tau}\right)$  (with duration  $\tau$ , pulse period  $T$  and peak intensity  $I_0 = \frac{\bar{I}T}{2\tau}$ ) [42], to obtain  $P(I) = \frac{\tau}{T} \frac{1}{I \sqrt{1 - \frac{I}{I_0}}}$ .

Considering the nonlinear mechanisms observed in our experiment, the spontaneous Four Wave Mixing efficiency (defined as the number of pairs generated per second) will be given by the second order moment  $\langle I^2 \rangle$ , whereas the Spontaneous Raman scattering will be related to the first order moment  $\langle I \rangle$  (in contrast to the case of the stimulated regime of Raman amplification treated in [39]). The calculation of these moments for the different regimes are summarized in Table 1. The main result is the confirmation of the expected observation of a two times better photon pair emission in the case of the multimode CW source for the Spontaneous Four Wave Mixing mechanism. We can note that this improvement is not expected in the case for the Spontaneous Parametric Down Conversion for the emission of photon pairs [43], or for the Third Order Spontaneous Parametric Down Conversion for the emission of photon triplets [44] as these mechanisms, similarly to the spontaneous Raman scattering, are governed by the first order moment.

When compared to experimental generation efficiencies one obtains a good accordance between the predictions of the model and the experiments, especially the better performance of the highly multimode CW Ti:Sapphire laser is correctly explained. Identically, the order of magnitude of the difference between the pulsed and the CW regime is also correctly evaluated. The slightly better than expected efficiency in the pulsed regime, might

be due to a better injection of the pump beam for that particular experiment illustrated by a slightly better transmission of the nanofiber (32% instead of 28% in the CW regime at the time of the experiment).

The expression in the pulsed regime shows that the efficiency is governed by the Peak – Average power product or said a different way that it is inversely proportional to the pulse duration. This last variation is only valid if all the photons are collected by the detection set-up, that can be a problem with very short pulses as the JSI is broadened inversely proportionally to the pulse duration [34]. In our experimental condition, this relation is still valid and was confirmed experimentally (see Insert in Fig. 2) on the short range of variation of the pulse duration allowed by the laser dispersion control system ( $\tau$  varying from 1 to 2 ps).

Table 1: Parameters for the evaluation of relative nonlinear efficiencies for a given mean intensity  $\bar{I}$ , for the single- and multi-mode CW sources and the pulsed sources delivering hyperbolic secant pulses (with a duration  $\tau = 1.03ps$  and a pulse period  $T = 12.5ns$ ).

Source characteristics	Intensity distribution $P(I)$	First order moment $\langle I \rangle$	Second order moment $\langle I^2 \rangle$	Raman Relative intensity	Theoretical Pair generation relative efficiency	Experimental Pair generation relative efficiency
CW Singlemode	$\delta(I - \bar{I})$	$\bar{I}$	$\bar{I}^2$	1	1	1
CW Multimode	$\frac{1}{\bar{I}} e^{-\frac{I}{\bar{I}}}$	$\bar{I}$	$2\bar{I}^2$	1	2	1.96
Sech – Pulsed	$\frac{\tau}{T} \frac{1}{I \sqrt{1 - \frac{I}{I_0}}}$	$\bar{I}$	$\frac{2 I_0 \bar{I}}{3}$ $= \frac{T}{3\tau} \bar{I}^2$	1	4050	5034

## V CAR Measurements

### V-A CAR in the different temporal regimes of the pump

To characterize the performances of the nanofiber as a source of pair of photons we need to characterize the Coincidence to Accidental Ratio (CAR). The coincidences were measured previously by counting the number of counts in the coincidence peak. The Accidentals are measured by counting the counts outside this coincidence peak. The exact measure depends on the temporal regime. In the pulsed regime, we clearly see (see insert in Fig. 4), in the coincidence histogram, peaks with a temporal separation equal to the repetition

period  $T$  of the laser. The noise excess in these periodic peaks is mainly due to occurrence between unpaired parametric photons of different pulses [15] (i.e a photon of a pair whose twin photon has not been detected in the other branch). We thus measure the mean number of counts in a large number of peaks (typically 300 on each side of the coincidence peak) on a temporal window equal to the one used to measure the number of counts in the coincidence peak (i.e.  $t_{bin} = 1.3ns$ ). In the CW regime, the measure is simpler as all time-bins outside coincidence peak contribute in the same manner to the noise. We then just measure the average number of counts on a large temporal window on both side of the coincidence peak, here again normalized on the same temporal window than the coincidence peak measurement. These measurement protocols allow to extract the measured parameter from a large number of samples, i.e. with a low error bar (calculated supposing a Poisson noise statistics), with moderate measurement duration (typically some tens of minutes and most of the time less than one hour) even in the case of very high CAR and low powers. For the experimental conditions of Figure 2, we measured a maximum value of the CAR of 7000 in the pulsed regime (Fig. 3), 2300 in the single mode CW regime and 2700 in the multimode CW regime. These values are of the order of magnitude of the best values observed in fibered structures [10, 15, 17], despite being still limited firstly by the Raman contribution on the idler side and secondly to a lower extent by the dark counts on the signal side.

### V-B Optimization of the CAR

To decrease Raman scattering and increase the CAR, we first choose to shift Ti:Sapphire pump wavelength to 839nm. The idler beam being shifted to 1550nm, the wavelength shift between pump and idler photons is thus increased by 30nm. This allows to go from a situation where we were in the vicinity of the 12<sup>th</sup> Raman peak of silica (at 440cm<sup>-1</sup>) to a situation where we pass this peak and are on the beginning of the leading edge of the much lower 13<sup>th</sup> Raman peak. The second improvement used for CW experiment (in the pulsed regime the source can still be considered as virtually Raman free) was to use a smaller width passband filter by replacing the CWDM filter by a tunable filter that reduces the almost white Raman Noise. We choose a filter with a width of 3nm centered on the central peak of the JSI. We lose some of the parametric pairs, due to this filtering (only 40% of the emitted spectra passes the filter, that would normally be able to detect the whole JSI peak if the fiber had been perfectly uniform) and because of the lower (around 40%) transmission of the filter,

but nevertheless obtained a better signal to noise ratio. These different improvements allow us to reduce the Raman counts by more than two orders of magnitude (Appendix E), without nevertheless completely eliminating it (the Idler counts still varying linearly with the pump power). We also benefit from the two times higher emission rate of pairs due to the multimode nature of the pump (no single mode laser diode was available at that wavelength for comparison). The signal, idler counts and coincidences rates have a similar behavior as in the previous measurement and results of polynomial adjustment of the power dependences are shown in Appendix E.

We firstly characterize the CAR in the CW regime, measured as a function of the generated pair numbers per bins (given by  $p = \frac{R_C}{\eta_S \eta_I} t_{bin}$  in the CW regime) still for a value  $t_{bin} = 1.3ns$  corresponding to the width of the coincidence peak. Considering that most of the detected counts on the detectors are in fact contributing to noise, the CAR can be identified (see blue/dark gray and green/light gray diamond in Fig. 4) to the normalized second order correlation defined as  $g_{SI}^{(2)} = \frac{R_C}{R_S R_I t_{bin}}$  in the CW regime [10]. This allows for an evaluation of the CAR by considering only the detectors counts and coincidences rates. CAR (and  $g_{SI}^{(2)}$ ) reaches a value of  $24000 \pm 2000$  at a pump power around 24mW and a generation rate around 5 kCounts/s (see Appendix F for an alternate analysis of  $g_{SI}^{(2)}$  and comparison with present state of the art).

We then characterize the CAR in the pulsed regime, the experimental set-up is almost identical as the one for Fig. 2 except that we use the 1550nm output of the CWDM filter (as the Raman noise is not as critical as in the CW regime), and a slightly longer pulse duration  $\tau = 1.40ps$  compared to the pump at 852nm. The results are presented in Fig.4 (blue/dark gray disks) as a function of the generated pair number per pulse (given here by  $p = \frac{R_C}{\eta_S \eta_I} T$ ). As expected from CAR model [15] CAR varies as  $p^{-1}$  for high  $p$ , until a saturation at low  $p$  due to the influence of dark counts. CAR is always higher than 100 even at the highest generation rate around 1 MCounts/s at 8mW power and reaches values as high as  $41000 \pm 6000$  for an average pump power around 100-150  $\mu W$ .

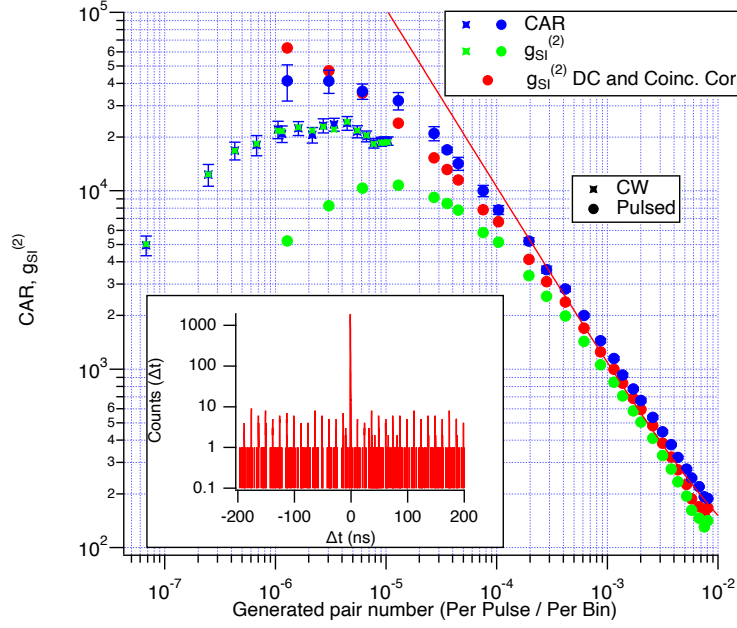


Figure 4: Measured CAR (blue/dark gray symbols) and calculated  $g_{SI}^{(2)}$  (green/light gray symbols) as a function of the generated number of pairs per pulse ( $p$ ) in the pulsed regime ( $\tau = 1.4$  ps) (disks) and per bins in the multimode CW regime (diamonds). The red/gray line is a guideline indicating a  $p^{-1}$  variation. The red/gray symbols represent the calculated  $g_{SI}^{(2)}$  corrected from the Dark Count contribution and from the coincidence contribution to the Signal and Idler count rates (see text for precisions). The pump wavelength was 839nm in both experiments, we estimated  $\eta_S = 9\%$  and  $\eta_I = 5\%$  in the pulsed regime and  $\eta_S = 8\%$  and  $\eta_I = 1\%$  in the CW regime the lower idler detection efficiency being due to the higher loss induced by the use of the tunable filter compared to the CWDM filter used in the pulsed experiment. The insert shows a typical coincidence histogram in the pulsed regime at a mean power of 5.13mW with a CAR of 450 (acquisition time 10s).

If we calculate the normalized second order correlation ( $g_{SI}^{(2)} = \frac{R_C}{R_S R_I T}$ ) adapted to the pulsed regime [17], we see that this parameter (green/light gray disks in Fig.4) clearly underestimates the CAR, both in the high pair number regime and in the low pair regime, even if in both cases the reason is different. In the high pair number regime,  $R_S$  and  $R_I$  that are used to estimate the accidental include a non-negligible amount of coincidence counts (all the more that the detection efficiency is high) that in practice do not participate to the accidental (as they are already counted as real pairs). These  $R_C$  have thus to be subtracted to the count rates  $R_S$  and  $R_I$  (or equivalently  $g_{SI}^{(2)}$  has to be divided by  $(1 - \eta_S)(1 - \eta_I)$  [15]). For low number of generated pairs, the underestimation is due to the influence of the Dark Counts that are implicitly supposed to fully contribute to the Accidental. This problem is usually solved by correcting  $R_S$  and  $R_I$  from the dark count rates [17] but in that case  $g_{SI}^{(2)}$  is shown to vary as  $p^{-1}$  [15] without saturation at low power (see the line in Fig. 4) and thus with a clear overestimation of the CAR. In fact, in the pulsed regime the dark count photons are dispatched on all time bins when the accidentals are calculated only for some time bins synchronous to



the pulse. Contrarily to [17], we thus choose to correct only partially for the dark count by taking into account the ratio of the accidental time measurement window  $t_{bin}$  to the repetition period  $T$ . The corrected expression that takes into account both these effects is:

$$g_{SI}^{(2)} = \frac{R_C}{\left(R_S - \left(1 - \frac{t_{bin}}{T}\right) DCR_S - R_C\right) \left(R_I - \left(1 - \frac{t_{bin}}{T}\right) DCR_I - R_C\right) T} \quad (3)$$

with  $DCR_I = 65s^{-1}$  and  $DCR_S = 53s^{-1}$  the dark count rate of the used detectors. It is shown as the red/gray disks in Figure 4 and shows a good accordance with experimentally measured CAR. The small residual difference between  $g_{SI}^{(2)}$  and CAR (red/gray and blue/dark gray circles in Fig. 4) is most probably due to the impact of detector after-pulses (estimated to  $P_{AP} \approx 10\%$  for the InGaAs detector at the used deadtime value [35] and  $P_{AP} \approx 0.5\%$  for the silicon detector) that causes an excess number of counts that will participate to the accidental (a complete analysis of their exact influence on the CAR has still to be realized).

## VI Conclusion

We have thus observed for the first time, generation of pair of photons in the CW regime in a nanofiber structure. We have observed that temporal fluctuations due to the multimode nature of the CW pump beam induce a two times excess of pair generation compared to a single mode source, that has been analyzed for the first time in the context of nonlinear quantum sources of photon pairs. This result is also important on the practical aspects as multimode source are common in the field on nonlinear optics. For example, the largely used pulsed Ti:sapphire laser can easily be operated in the CW regime by stopping mode-locking, which allows to perform a rapid and direct comparison of the CW and the pulsed regime that nevertheless needs to consider the multimode nature of its CW operation for a correct comparison. The use of multimode source is also interesting for future development in an application point of view as these sources are usually more powerful, easier to use and cheaper than single mode lasers.

The nanofiber geometry is very interesting as the nanofiber can be pulled with large length and controlled diameter that allow to control phase matching properties and emission efficiency. The nanofiber is pulled together with adiabatic tapers that allow to assure almost lossless injection from the access fiber mode to the nanofiber mode. The obtained results are very promising considering the relatively small length (1cm) of the nanofiber, and we have

shown that such a nanofiber can be pulled with a relatively good uniformity, with diameter fluctuations of about  $\pm 2$  nm, sufficient to observe a close to ideal phase matching curve shape. Controlling the nanofiber diameter on longer length will be the next objective of the study and first results on the reproducibility of the nanofiber characteristics [30], make us feel confident in reaching that objective.

Finally, the realized source presents very good performances with pairs emission rates that exceed 1Mcounts/s in the pulsed regime and more than 1kcounts/s in the CW regime for moderate pump power in the 10mW range. Even if the performances are still limited by spontaneous Raman scattering (operated in the vicinity of the 12<sup>th</sup> Raman order), the source also presents CAR values largely above several thousands, with optimized maximum values of 41000 and 24000 in the Pulsed and CW multimode regimes respectively. These CAR values measured in the CW and pulsed regimes are, to the best of our knowledge, among the highest measured in a fiber structure geometry. This makes us confident in the possibility to observe even higher CAR in architectures where Raman scattering is reduced such as in fibers operated at low temperature [8] and used with improved superconducting detectors [10].

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## Appendix A. Joint Spectral Intensity

The biphoton state created by a pump at frequency  $\omega_p$  at the output of the nonlinear media usually writes, in the low efficiency regime, as [28, 45, 34]:

$$|\psi(t)\rangle = |0, 0\rangle + \iint JSA(\omega_p, \omega_s, \omega_l) |1_{\omega_s}, 1_{\omega_l}\rangle d\omega_s d\omega_l \quad (A1)$$

where the Joint Spectral Amplitude (JSA) characterizes the emission spectra of the emitted pair, and its square modulus ( $JSI(\omega_p, \omega_s, \omega_l) = |JSA(\omega_p, \omega_s, \omega_l)|^2$ ) the Joint Spectral Intensity represents the probability of emission of a pair in a given spectral domain.

This JSA (or JSI), except for a slowly frequency dependent term related the nonlinear efficiency of the medium ( $C$  that will determine the efficiency of pair generation and will

depend on multiple parameters including pulse energy and duration), is given by a product of two normalized functions related to energy and momentum conservation (see Fig. 5):

$$JSA(\omega_p, \omega_s, \omega_I) = C g(\omega_p, \omega_s, \omega_I) \times f(\omega_p, \omega_s, \omega_I) \quad (A2)$$

The energy conservation term depends on nonlinearity order and for Spontaneous Four Wave Mixing it is given by  $g(\omega_p, \omega_s, \omega_I) = [\tilde{G} * \tilde{G}](\omega_s + \omega_I - 2\omega_p)$ , with  $\tilde{G}(\omega) = FT[G(t)]$  the Fourier Transform of the normalized temporal shape  $G(t)$  of the incident pulse amplitude. For the hyperbolic-secant pulse considered here  $G(t) = sech(t/\tau)$ , an analytical expression for the normalized pulse autocorrelation spectra can be found  $[\tilde{G} * \tilde{G}](\omega) = (\pi\tau\omega/2)/\text{Sinh}(\pi\tau\omega/2)$ . The width of this function (Fig. 5-a) depends on the pulse duration becoming thinner when pulse duration increases (moving toward a Dirac function for a perfectly monochromatic pump). In the multimode CW regime, the multiple incoherent modes again broaden this function but without benefiting from the increasing of pulse peak power and then from the better efficiency brought by the phase locking of the mode in the pulsed regime.

The momentum conservation term  $f(\omega_p, \omega_s, \omega_I) = \frac{1}{L} \int_0^L e^{-i \Delta\beta(\omega_p, \omega_s, \omega_I) z} dz$ , depends on the phase mismatch of the nonlinear mechanism  $\Delta\beta(\omega_p, \omega_s, \omega_I)$  and simplifies in a uniform nonlinear medium of length  $L$ , to the well-known phase matching expression  $f(\omega_p, \omega_s, \omega_I) = e^{-i \Delta\beta \frac{L}{2}} \text{sinc}\left(\frac{\Delta\beta L}{2}\right)$ .

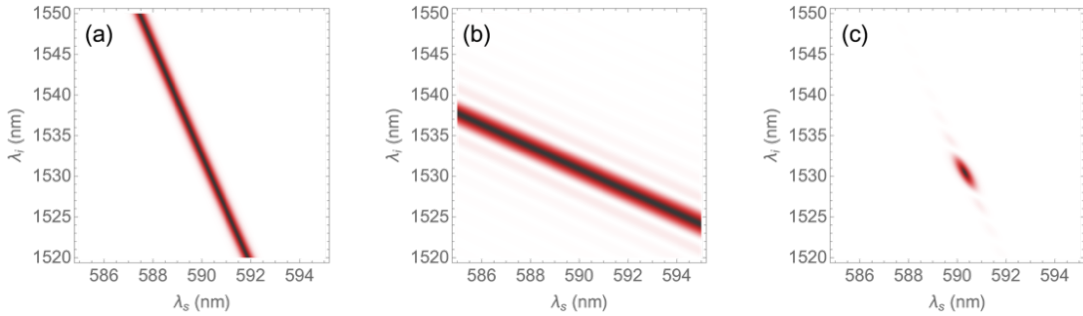


Figure 5: Example of a plot of  $|g(\omega_p, \omega_s, \omega_I)|^2$  (a), of  $|f(\omega_p, \omega_s, \omega_I)|^2$  (b), and of their product (c) giving  $JSA(\omega_p, \omega_s, \omega_I) = |JSA(\omega_p, \omega_s, \omega_I)|^2$  for a given pump wavelength. The calculation uses the typical parameters of the experiment.

## Appendix B. Nanofiber propagation properties simulations

The nanofiber is some kind of textbook object [46], it is perfectly theoretically described by a step index fiber model with a core of radius  $\rho$  and index  $n_{silica}$  surrounded by an infinite layer of air with index  $n_{Air}$ . The effective index  $n_{eff}$  of the fundamental mode of the nanofiber, i.e. the  $HE_{11}$  vectorial mode considered here, is given by the eigenvalue

equation (the equation and the described calculation can be easily generalized to any other propagation mode) [46] :

$$\left( n_{eff} \frac{V^2}{U^2 W^2} \right)^2 = \left( n_{Silica}^2 \frac{J_1(U)}{U J_0(U)} + n_{Air}^2 \frac{K_1(W)}{W K_0(W)} \right) \left( \frac{J_1(U)}{U J_0(U)} + \frac{K_1(W)}{W K_0(W)} \right) \quad (B1)$$

With  $V^2 = U^2 + W^2$ , and  $U^2 = \rho^2 \left( \frac{2\pi}{\lambda} \right)^2 (n_{Silica}^2 - n_{eff}^2)$  and  $W^2 = \rho^2 \left( \frac{2\pi}{\lambda} \right)^2 (n_{eff}^2 - n_{Air}^2)$ .

This equation can be reduced, still for the HE<sub>11</sub> vectorial mode, as:

$$f(n_{eff}, \lambda) = x_r + b_r(1 - \Delta) + \sqrt{b_r^2 \Delta^2 + \frac{c_r}{n_{Silica}^2}} = 0 \quad (B2)$$

With  $x_r = \frac{J_1(U)}{U J_0(U)}$ ,  $b_r = \frac{K_1(W)}{W K_0(W)}$  and  $c_r = \left( n_{eff} \frac{V^2}{U^2 W^2} \right)^2$ , using  $\Delta = \frac{n_{Silica}^2 - n_{Air}^2}{2n_{Silica}^2}$ . With wavelength independent core and cladding refractive indices the equation is solved as a function of the reduced variables U and V and the effective index is deduced, with a simple relation to the nanofiber radius and a general solution valid whatever this radius.

When dispersion of the different media ( $n_{Silica}(\lambda)$  [47] and even  $n_{Air}(\lambda)$  [48]) are taken into account, we lose this simple relation and the effective index wavelength dependence has to be calculated for each radius, by solving the implicit equation (B2) for the variable  $(n_{eff}, \lambda)$ . The equation is solved numerically for some specific points related to the zeros of a Chebyshev polynomial and a development of  $n_{eff}(\lambda)$  (or of the propagation constant  $\beta(\omega) = \frac{\omega}{c} n_{eff} \left( \frac{2\pi c}{\omega} \right)$ ) on the basis of Chebyshev polynomials is then realized and used for further calculations (Fig. 6).

In addition to give a way to numerically simulate the performance of the nanofiber, the polynomial expression of  $\beta(\omega)$ , allows to obtain simple analytical expressions for the quantity allowing to describe more completely the nanofiber behavior when some parameters are varied (such as for example in the case of a fluctuation of the radius).  $\beta(\omega)$  can be rewritten around a mean frequency  $\omega_{X0}$  (with X=P, S or I):

$$\beta(\omega_X) = \beta_{X0}^{(0)} + (\omega_X - \omega_{X0}) \beta_{X0}^{(1)} + \sum_{k=2}^N \frac{(\omega_X - \omega_{X0})^k}{k!} \beta_{X0}^{(k)} \quad (B3)$$

where  $\beta_{X0}^{(k)} = \frac{\partial^k \beta}{\partial \omega^k}(\omega_{X0})$ , and N is the order of the polynomial used to describe  $\beta(\omega)$  (we use the fact that  $\frac{\partial^{N+1} \beta}{\partial \omega^{N+1}}(\omega) = 0$ ).

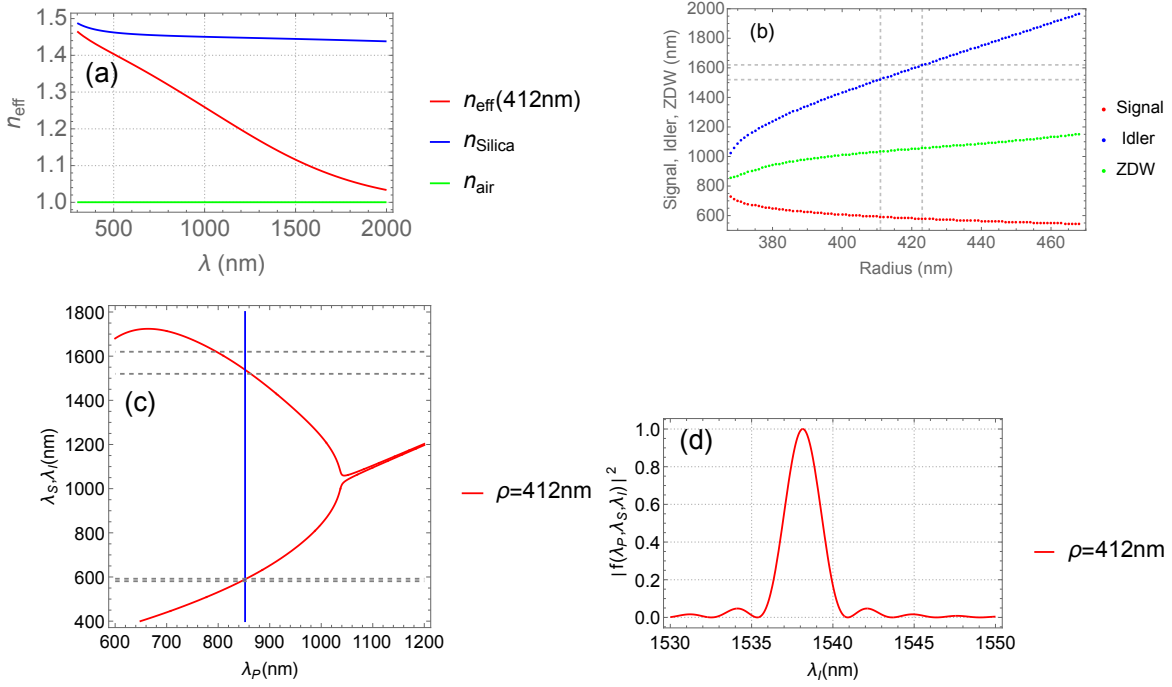


Figure 6: (a) Simulated value of the fundamental HE11 mode effective index for a nanofiber radius of 412nm.  $n_{\text{eff}}$  varies from the silica refractive index at wavelength where the mode is highly confined in the nanofiber core, to the air refractive index when the mode is deconfined. (b) From the effective index we can calculate the Zero Dispersion wavelength as a function of the nanofiber radius (green/light gray curve) and by solving the energy ( $\omega_{S0} - \omega_{I0} - 2\omega_{P0} = 0$ ) and momentum ( $2\beta(\omega_{P0}) - \beta(\omega_{S0}) - \beta(\omega_{I0}) = 0$ ) conservation equation simultaneously we can calculate for each radius the emitted Signal (red/red gray curve) and Idler (blue/dark gray curve) wavelength values as a function of radius for a pump wavelength of 852,2nm. This allows to determine the radius range allowing emission of idler photons in the telecom band (gray dashed lines). (c) the same calculation can also be performed for a fixed radius (here 412nm) as a function of pump wavelength, to evaluate tolerance on pump wavelength. (d) Finally, we can calculate the shape of emission spectra by tracing  $\text{Sinc}^2[(2\beta(\omega_{P0}) - \beta(\omega_S) - \beta(\omega_I))L/2]$  for a nanofiber of length  $L=1\text{cm}$  at energy conservation.

We can then calculate  $\Delta\beta(\omega_P, \omega_S, \omega_I) = 2\beta(\omega_P) - \beta(\omega_S) - \beta(\omega_I)$  and obtain, around the wavelength of perfect phase matching (i.e. for  $2\beta_{P0}^{(0)} - \beta_{S0}^{(0)} - \beta_{I0}^{(0)} = 0$  and  $2\omega_{P0} = \omega_{S0} - \omega_{I0}$ ):

$$\begin{aligned}
\Delta\beta(\omega_P, \omega_S, \omega_I) &= (\omega_S - \omega_{S0}) (\beta_{P0}^{(1)} - \beta_{S0}^{(1)}) - (\omega_I - \omega_{I0}) (\beta_{P0}^{(1)} - \beta_{I0}^{(1)}) \\
&+ 2 \sum_{k=2}^N \frac{(\omega_P - \omega_{P0})^k}{k!} \beta_{P0}^{(k)} - \sum_{k=2}^N \frac{(\omega_S - \omega_{S0})^k}{k!} \beta_{S0}^{(k)} \\
&- \sum_{k=2}^N \frac{(\omega_I - \omega_{I0})^k}{k!} \beta_{I0}^{(k)}
\end{aligned} \tag{B4}$$

which generalizes at all dispersion orders the first order expression  $\Delta\beta_1(\omega_P, \omega_S, \omega_I) = (\omega_S - \omega_{S0}) (\beta_{P0}^{(1)} - \beta_{S0}^{(1)}) - (\omega_I - \omega_{I0}) (\beta_{P0}^{(1)} - \beta_{I0}^{(1)})$  usually found in the literature [45] (we

can note that the high order terms for the pump can be considered as negligible as usually the pump spectral width can be considered as small enough for the dispersion influence to be limited to the group velocity  $v_g(\omega_{p0}) = \frac{1}{\beta_{p0}^{(1)}}$  term).

We can even go further, if we note that the ZDW and thus the emitted wavelength depends on the nanofiber radius. At first order for a small amplitude of the radius fluctuation, the dependence is linear (see Fig 6-b) and the linear shift of the radius will also linearly control the dispersion curve and the emitted wavelength shift. If the Zero Dispersion frequency is shifted by a quantity  $\Delta\omega_{ZDW}$  the new propagation constant  $\beta^S(\omega)$  of the dispersion shifted waveguide, will be given by:

$$\beta^S(\omega) = \beta(\omega - \Delta\omega_{ZDW}) = \beta(\omega) - \Delta\omega_{ZDW} \frac{\partial\beta}{\partial\omega}(\omega) \quad (B5)$$

We will then be able to write the phase mismatch at first order of  $\Delta\omega_{ZDW}$ , as:

$$\Delta\beta^S(\omega_p, \omega_s, \omega_I, \Delta\omega_{ZDW}) = \Delta\beta(\omega_p, \omega_s, \omega_I) + \Delta\omega_{ZDW} d\Delta\beta(\omega_p, \omega_s, \omega_I) \quad (B6)$$

with

$$d\Delta\beta(\omega_p, \omega_s, \omega_I) = 2 \frac{\partial\beta}{\partial\omega}(\omega_p) - \frac{\partial\beta}{\partial\omega}(\omega_s) - \frac{\partial\beta}{\partial\omega}(\omega_I) \quad (B7)$$

Using the relation  $\frac{\partial\beta}{\partial\omega}(\omega_X) = \beta_{X0}^{(1)} + \sum_{k=2}^N \frac{(\omega_X - \omega_{X0})^{k-1}}{(k-1)!} \beta_{X0}^{(k)}$ , we finally obtain:

$$\begin{aligned} d\Delta\beta(\omega_p, \omega_s, \omega_I) &= 2\beta_{P0}^{(1)} - \beta_{S0}^{(1)} - \beta_{I0}^{(1)} + (\omega_s - \omega_{S0}) (\beta_{P0}^{(2)} - \beta_{S0}^{(2)}) \\ &+ (\omega_I - \omega_{I0}) (\beta_{P0}^{(2)} - \beta_{I0}^{(2)}) + 2 \sum_{k=3}^N \frac{(\omega_p - \omega_{P0})^{k-1}}{(k-1)!} \beta_{P0}^{(k)} \\ &- \sum_{k=3}^N \frac{(\omega_s - \omega_{S0})^{k-1}}{(k-1)!} \beta_{S0}^{(k)} - \sum_{k=3}^N \frac{(\omega_I - \omega_{I0})^{k-1}}{(k-1)!} \beta_{I0}^{(k)} \end{aligned} \quad (B8)$$

The fluctuation of the Zero Dispersion frequency governs the emission spectra at first order through the term  $d\Delta\beta_1(\omega_p, \omega_s, \omega_I) = 2\beta_{P0}^{(1)} - \beta_{S0}^{(1)} - \beta_{I0}^{(1)} + (\omega_s - \omega_{S0}) (\beta_{P0}^{(2)} - \beta_{S0}^{(2)}) + (\omega_I - \omega_{I0}) (\beta_{P0}^{(2)} - \beta_{I0}^{(2)})$

### Appendix C. Characterization with Stimulated Emission Tomography (SET)

Characterization with a SET experiment allows to precisely locate the emission peak [33, 34], through access to the JSI (see Appendix A). For the SET measurement, a tunable

spectrally thin seed beam (on the idler side in our case) is injected together with the pump beam, this beam is amplified through stimulated four wave mixing, and simultaneously each amplified seed photon is accompanied by a twin photon on the signal side. Usually, the spectrum of these signal photons, corresponding in practice to a slice of the JSI (Fig. 5-c) at the used seed wavelength, is measured with a spectrometer. The seed wavelength is then varied, and a new spectrum is registered, allowing to finally reconstruct the JSI (insert of Fig. 7-b). Considering the typical spectral width of the pulsed pump beam (typically around 0.25nm, with largest values at 0.5nm for shortest pulses (Fig. 7-a)) and the limited resolution of the used fibered spectrometer (Ocean Optics USB2000) around 1nm (See insert in Fig. 7-b), we can hardly resolve the energy conservation contribution to the JSI, and the response is mainly governed by the phase matching function at energy conservation, which has the well-known Sinc shape. That's why we prefer to adapt the classical SET set-up, by replacing the spectrometer directly by the single photon detector of our experiment, used with a 10nm interferential band pass filter positioned around the expected response wavelength (it would correspond to integrate the whole spectral response of the spectrometer for each value of the idler wavelength, similar to a projection of the JSI (Fig. 5-c) on the idler wavelength axis (and converted to the signal wavelength using energy conservation relation). Due to the very high sensitivity of the single photon detectors, we can then obtain JSI at low power and even in the CW regime with high dynamic (the only constraints being that the stimulated response has to overpass the spontaneous one), without losing the spectral resolution given by the much smaller spectral width and by the tuning step of the tunable seed. The tunable laser diode wavelength is tuned at constant speed and fixed wavelength steps, and the temporal variation of the single photon detector Count Rate is registered, the temporal step being related to the tuning speed, a precise wavelength SET response curve can then be registered. Figure 7-b shows a typical spectrum we obtained with a pump at 852nm. We observed no difference in the spectral shape obtained in the pulsed and CW pump regime (with a typical average power in the mW range), indicating negligible influence of SPM pulse broadening in our experimental conditions [49]. The JSI shows a three peaks structure (Fig. 7-b), with a main peak width close to the spectral width expected for our 1cm nanofiber length (see Fig. 6-d). We attribute this JSI broadening to nanofiber diameter fluctuations that cause fluctuations of the Zero Dispersion Wavelength (ZDW) of the nanofiber. Usually, these fluctuations are random, and the peak shape is strongly affected with high chaotic broadening [36, 50, 10] that

most of the time prevents to reversely determine the fiber fluctuation shape from the JSI shape. In our case, the rather organized shape of the JSI allows to deduce that our nanofiber presents probably a sinusoidal fluctuation of its diameter (Appendix D).

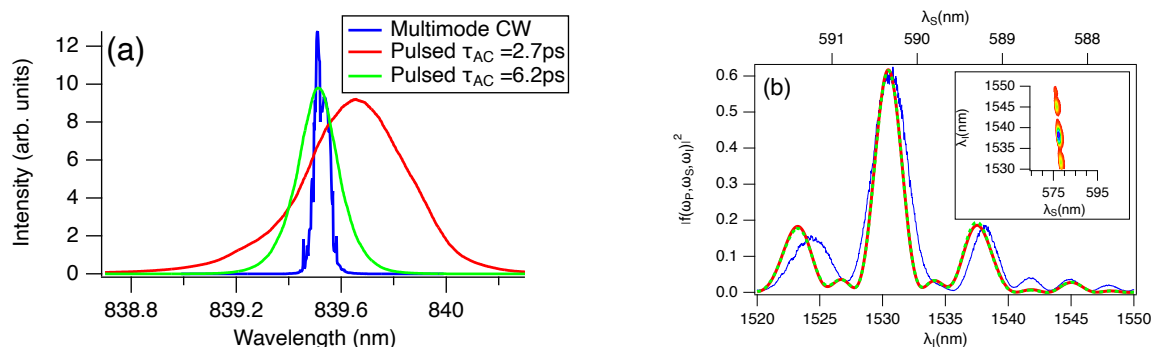


Figure 7: (a) Typical spectrum of the Ti:Sapphire laser in the multimode CW regime, and in the pulsed regime (at two extremal pulse autocorrelation durations). In the CW regime it is only slightly thinner than in the pulsed regime and would give a JSI with a similar width. The single mode CW laser diode is not shown, as its much thinner spectrum cannot be resolved by our Optical Spectrum Analyzer. (b) Experimental measurement of the photon pairs emission spectra at energy conservation for a pump at 852nm,  $\lambda_I$  is the tunable laser diode wavelength and  $\lambda_S$  is determined through energy conservation (or by the fibered spectrometer calibration) giving function  $|f(\omega_p, 2\omega_p - \omega_I, \omega_I)|^2$  (blue/dark gray curve). Comparison with simulation using a sinusoidal fluctuation of the ZDW along propagation, with period  $z_p = 3.83\text{ mm}$  an amplitude of fluctuation  $\Delta\lambda_{ZDW} = 2.2\text{ nm}$  and a phase  $\phi_0 = -2\text{ rad}$ , dashed green/light gray curve represents the calculation with analytical expression (D2), whereas red/gray curve represent the numerical model of [36]. The insert shows a classical SET spectral map obtained with a fibered spectrometer for a slightly different pump wavelength (around 840nm) and a slightly more stretched nanofiber. The JSI is characteristics of a correlated state expected for a general phase matching condition in which group velocity matching is not optimized [34]. The spectral width of the JSI along the signal wavelength axis is limited by the spectral resolution of the used fibered spectrometer and is thus the same for all type of pump.

These preliminary results allow to optimize the detection set-up. Indeed, we can see that the spectral extension of the emitted photons is mostly in a 20nm band around 1530nm (and a 3nm band around 590nm on the signal side). These bands are well covered by the 1530nm spectral channel of our commercial CWDM filter (and by a 590nm band pass filter of 10nm bandwidth). This means that almost all the emitted photons pairs can be efficiently detected. We can also note that the main peak width is around 3nm and that a better filtering of noise is possible at the expense of a moderate loss of coincidences (as only the lateral peaks are lost). Furthermore, we can also accommodate for small fluctuation of the pulsed laser pump wavelength, that we estimate we can control with an accuracy of the order of  $\pm 0.3\text{nm}$  depending on precise adjustment of the laser cavity on a day-to-day basis, as well as compared to the much more stable CW laser diode frequency (these fluctuations cause an idler wavelength fluctuation of less than  $\pm 0.5\text{nm}$  that is largely compatible with the spectral width of our filters).



## Appendix D. Determination of the nanofiber shape

To deduce the nanofiber longitudinal shape, we use the expression of the phase matching function in presence of ZDW fluctuation, given for a fiber of length  $L$ , by [50]:

$$f(\omega_P, \omega_S, \omega_I) = \frac{1}{L} \int_0^L dz e^{i \int_0^z \Delta\beta^S(\omega_P, \omega_S, \omega_I, z') dz'} \quad (D1)$$

where  $\Delta\beta^S(\omega_P, \omega_S, \omega_I, z)$  is the position dependent phase mismatch between the different propagating waves. We have to note here that the relation (D1) can be used to calculate the effect on the JSI of a waveguide fluctuation, but that, as the JSI corresponds to an integration of the whole waveguide response, it is not possible to reverse it uniquely and obtain the fluctuation directly from the JSI spectrum. Hypotheses must be made on possible cause of fluctuations knowing the characteristics of the waveguide (for example in our case, a possible contribution of parts of the tapers instead of the nanofibers or different shapes of fluctuations of the nanofiber) then some signatures of these fluctuations can be searched in the measured JSI, to arbitrate between the different possibilities. For the studied fiber, we finally opt for the simple sinusoidal fluctuation, that gives good accordance with a small number of free parameters, always remembering that more complex fluctuations might probably give similar JSI at the expense of a larger number of free parameters.

In the general cases, the analysis of this relation is based on numerical models [36] cutting the fiber in small pieces. Nevertheless, in some special cases, such as for a sinusoidal fluctuation, an analytical development can be made.

We write  $\Delta\beta^S(\omega_P, \omega_S, \omega_I, z) = \Delta\beta(\omega_P, \omega_S, \omega_I) + \Delta\omega_{ZDW}(z) d\Delta\beta(\omega_P, \omega_S, \omega_I)$ , with the position dependent zero dispersion frequency  $\Delta\omega_{ZDW}(z) = \Delta\omega_{ZDW0} \sin\left(\frac{2\pi z}{z_p} + \phi_0\right)$ , and with  $\Delta\beta(\omega_P, \omega_S, \omega_I)$  and  $d\Delta\beta(\omega_P, \omega_S, \omega_I)$  given by Eqs. (B4) and (B8) respectively (their implicit frequency dependence will be omitted in the following to simplify notations). The fluctuation is then quantified by only three parameters: the amplitude variation of the zero dispersion wavelength  $\Delta\lambda_{ZDW0}$  around the mean value  $\lambda_{ZDW}$  of the reference uniform diameter nanofiber, giving  $\Delta\omega_{ZDW0} = \frac{2\pi c \Delta\lambda_{ZDW0}}{\lambda_{ZDW}^2}$  (with  $c$  the light celerity), the period  $z_p$  of the fluctuation, and a phase  $\phi_0$  related to the exact position of the fluctuation on the nanofiber. We obtain finally after some calculations:

$$f(\omega_P, \omega_S, \omega_I) = i e^{i \left( \frac{\Delta\beta L}{2} + \frac{\Delta\omega_{ZDW0} d\Delta\beta z_p}{2\pi} \cos(\phi_0) \right)} \times \sum_{n=-\infty}^{n=+\infty} e^{i n \left( \frac{\pi L}{z_p} + \phi_0 + \frac{\pi}{2} \right)} J_n \left( -\frac{\Delta\omega_{ZDW0} d\Delta\beta z_p}{2\pi} \right) \text{Sinc} \left( \frac{\Delta\beta L}{2} + \frac{\pi n L}{z_p} \right) \quad (\text{D2})$$

In first approximation (i.e. for small fluctuations), the JSI will present side peaks having the usual  $\text{Sinc} \left( \frac{\Delta\beta L}{2} \right)$  shape. The peaks amplitude is governed by the Bessel function  $J_n$  of order  $n$ , with an argument directly related to the product of the amplitude of the fluctuation with the period of the fluctuation. This means that the shorter the fluctuation period, the higher the tolerable amplitude fluctuation. The peaks positions are directly related to the fluctuation period  $z_p$  (with the condition  $\Delta\beta z_p = 2\pi n$ , meaning that the peak would be at the zeros of the phase matching curve of an identical perfect nanofiber of length  $z_p$ ). Evidently, for high fluctuations, the different terms contributions mix and the JSI shape will become more chaotic due to the influence of the different phase terms.

If we restrict to the first three orders of the development (0 and  $\pm 1$ ), we find with this model the three observed peaks (Fig. 7-b). This response is governed by the dispersion characteristics of the nanofiber. In the simplest model of dispersion [45] using first order Taylor expansion of the dispersion around the perfect phase matching frequencies (see Appendix B) we have simply  $\Delta\beta = (\omega_S - \omega_{S0}) (\beta_{P0}^{(1)} - \beta_{S0}^{(1)}) - (\omega_I - \omega_{I0}) (\beta_{P0}^{(1)} - \beta_{I0}^{(1)})$  and  $d\Delta\beta = 2\beta_{P0}^{(1)} - \beta_{S0}^{(1)} - \beta_{I0}^{(1)} + (\omega_S - \omega_{S0}) (\beta_{P0}^{(2)} - \beta_{S0}^{(2)}) + (\omega_I - \omega_{I0}) (\beta_{P0}^{(2)} - \beta_{I0}^{(2)})$ , where  $\beta_x^{(k)} = \frac{\partial^k \beta}{\partial \omega^k}(\omega_x)$  are known. This allows rapid determination of the fluctuation parameters. Nevertheless, we use in our simulations, as usually done in the literature, higher order terms in the Taylor development of the dispersion (and larger number of peaks in equation (D2)) that gives a little more precise calculation (but without changing fundamentally the behavior) at the expense of more cumbersome expression and less physical insight of the significance of different terms. The spectral structure of the emission spectra can be rather well described by that simple model with a sinusoidal fluctuation of the ZDW with a period of 3.83mm (around the reference value of  $\lambda_{ZDW} = 1036\text{nm}$ ) and an amplitude of 2.2nm. Such a variation of the ZDW corresponds to a variation of the diameter of the nanofiber of about  $\pm 2$  nm (Fig. 6-b), around the mean diameter of 824nm. The origin of this fluctuation is currently unknown

probably it is due to variations of pulling parameters, it may also be due to residual strains in the nanofiber that may cause change of the ZDW. Moreover, if the sinusoidal fluctuation gives a good idea of the emission spectra the accordance is not perfect and the exact form is probably different, all the more than other periodic fluctuation (square, triangular, ...) can give spectrum with similar shapes. Nevertheless, these results show that our nanofiber is relatively uniform and show an emission spectrum rather close to the ideal Sinc shape.

## Appendix E. Counts and Coincidence data adjustment parameters

The experimental points presented in Fig. 2, were adjusted using a third order polynomial law  $R_X = a_X + b_X \bar{I} + c_X \bar{I}^2$ , ( $X = S, I, C$ , for Signal, Idler and Coincidence) as a function of the pump beam mean power  $\bar{I}$ . The adjustment uses Orthogonal Distance Regression (ODR) method of fit proposed by our data treatment software IgorPro. The adjustment takes into account the error bars on the experimental data and imposes the adjustment coefficients to have a physical significance and thus to be positive.

For the detector count rates, the constant parameter  $a_X$  corresponds to the dark count rates that is measured independently and subtracted from the experimental points, so we have  $a_S$  and  $a_I$  that can be considered to be zero. For the coincidences rate, the expected constant parameter  $a_C$  related to dark count accidental coincidences is very small and in practice not measurable in the condition of the presented experiment. It was kept in the adjustment and systematically found to be zero (remembering that we impose this parameter to be positive).

Thus, the only relevant parameters of the adjustment are thus the linear  $b_X$  and quadratic  $c_X$  parameters, the value of these parameters extracted from the data of Fig. 2 are presented in Table 2.

Table 2: Adjustment parameters of the quadratic polynomial fit of experimental data of Fig. 2. The parameters in italic correspond to parameters determined as close to or smaller than their errors bars. The parameters in bold are the ones used to calculate experimental data in Table 1.

Source characteristics	Count type	Adjustment linear coefficient $b_x (mW^{-1}s^{-1})$	Adjustment Quadratic coefficient $c_x (mW^{-2}s^{-1})$
CW Singlemode	Signal	<i>0.21 ± 0.25</i>	0.532 ± 0.034
	Idler	<b>1031 ± 14</b>	<i>0.83 ± 2.2</i>
	Coincidence	<i>0 ± 0.017</i>	<b>0.0291 ± 0.0028</b>
CW Multimode	Signal	<i>1.72 ± 0.44</i>	1.06 ± 0.06
	Idler	<b>1088 ± 14</b>	<i>0 ± 2.35</i>
	Coincidence	<i>0.0012 ± 0.027</i>	<b>0.0570 ± 0.0044</b>
Sech – Pulsed	Signal	<i>0 ± 58</i>	2850 ± 43
	Idler	<b>1183 ± 41</b>	1070 ± 20
	Coincidence	<i>0 ± 13</i>	<b>146.5 ± 3.4</b>

Some of the parameters in the table are much smaller than their error bars, these parameters can thus be considered as not relevant. This may correspond to a contribution that can reasonably be considered as non-existing, such as for example, the linear coefficient of the coincidences rates. This can also be a parameter that corresponds to a minor contribution compared to the other one in the conditions of the experiment. This is illustrated in Figure 8-a,b, where we see that the quadratic component of the Idler is almost not visible or for the signal where the linear component is much smaller than both quadratic and dark count ones. In both cases, the numerical adjustment can find a value for the corresponding parameters, but we cannot be confident in the found value. Some cases are less clear, for example the Idler count rate in the pulsed regime in Figure 8-c, where the linear contribution is only visible at low power and would have probably required more experimental points in that range to decrease the error bar.

The Signal, Idler counts and coincidences rates parameters associated to the CAR data of Figure 4, are shown in Table 3, we have similar but slightly lower performance compared to experiment at 852nm, probably because of a less optimized experiment and a longer pulse duration. We can note that the Raman contribution (measured by the Idler count rate), is almost two orders of magnitude smaller, thanks to a larger wavelength shift and a better noise filtering.

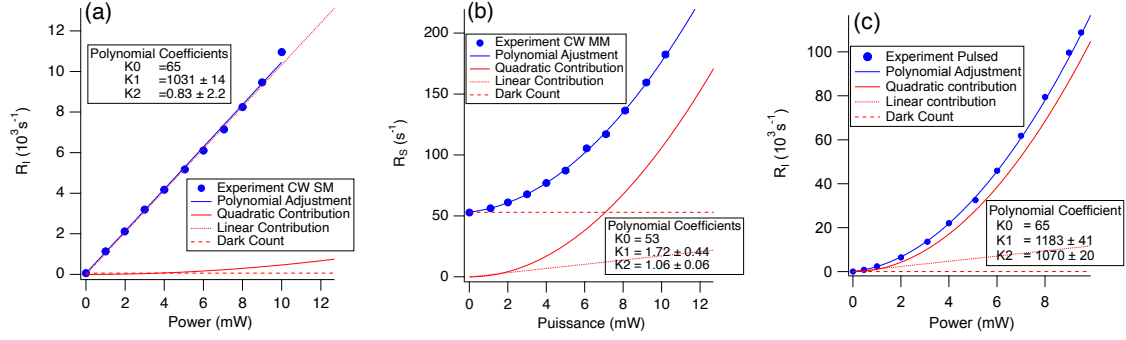


Figure 8: Polynomial adjustment of the Idler (a) experimental count rate of the Single mode CW regime and Signal (b) experimental count rate of the Multimode CW regime, with shown the amplitude of the contribution of the different constant (i.e Dark Counts), linear and quadratic terms. Contrarily to Fig.2, the dark count rate had not been subtracted and the curves are presented in linear scales. For Idler (a), the adjustment shows a small quadratic contribution much smaller than its error bar. The term is considered as not relevant as completely masked by the linear contribution. For the Signal (b), the roles are exchanged, the linear contribution that is found by adjustment has a large error bar and is considered as irrelevant, as it is completely masked either by the quadratic term (at high power) or by the Dark Count term (at low power). The main contribution is the quadratic term that is precisely determined, even though its value seems comparable to the equivalent term of the Idler. In the case of the idler count rate in the pulsed regime (c), we are in a mixed situation, the linear term can be considered as relevant as it is of the order of the quadratic term at low power but stays higher than dark count term, but the small number of experimental data in that region where this term dominates probably increases the error bar of the determined coefficient.

Table 3: Adjustment parameters of the quadratic polynomial fit of the signal, Idler counts and Coincidences rates related to the experimental data of Fig. 4. Note that, contrary to data of Table 2, the experimental condition had slightly changed between the two experiments, making the direct comparison of CW and pulsed regime more difficult. Taking into account the detection efficiencies, the generation rates are, in the pulsed regime  $\mu_P = 9.5 \times 10^3 \text{ pairs.s}^{-1}\text{mW}^{-2}$ , and in the CW regime  $\mu_P = 6 \text{ pairs.s}^{-1}\text{mW}^{-2}$ , slightly lower than with the pump at 852nm probably due to a less well optimized experimental set-up.

Source characteristics	Count type	Adjustment linear coefficient $b_x \text{ (mW}^{-1}\text{s}^{-1}\text{)}$	Adjustment Quadratic coefficient $c_x \text{ (mW}^{-2}\text{s}^{-1}\text{)}$
CW Multimode	Signal	$0 \pm 0.07$	$0.486 \pm 0.007$
	Idler	$9.69 \pm 0.11$	$0 \pm 0.0046$
	Coincidence	$0 \pm 0.012$	$0.0047 \pm 0.0001$
Sech – Pulsed	Signal	$0 \pm 3.1$	$877 \pm 8$
	Idler	$389 \pm 5$	$378 \pm 5$
	Coincidence	$0.68 \pm 0.51$	$43.32 \pm 0.49$

## Appendix F. Alternate analysis of the $g_{SI}^{(2)}$ and comparison to present state of the art

The Coincidence to Accidental Ratio (CAR) values measured in the CW and pulsed regime are, to the best of our knowledge, the highest measured in a fibered structure geometry. This is all the more true than we consider a rather conservative analysis procedure

especially for the CW regime. Indeed, we use a value of  $t_{bin}$  that allows to cover the whole coincidence peaks, but also gives a higher value for the noise counts (in the pulsed regime, the accidentals are, except perhaps at the lowest power, governed by peaks with similar shape compared to the coincidence peak so the value taken for  $t_{bin}$  is less critical).

To illustrate this point and compare our results to data that presently represents state of the art in similar structures, we use for the CW regime a data treatment similar to the one used in ref. [10] i.e. we plot  $g_{SI}^{(2)}(\Delta t) = \frac{R_C(\Delta t)}{R_S R_I t_{bin}}$  with a  $t_{bin}$  value given by the temporal resolution of the coincidence measurement device (100ps in our case) (see Fig. 9). This treatment would have given a peak value of 65000, i.e. around 2.7 times better than the maximum value of the CAR (or  $g_{SI}^{(2)}$ ) of 24000 we measured for the same set of experimental data. This better signal to noise ratio is obtained at the expense of a smaller number of detected pairs as the coincidence peak as a larger width and some coincidences are then outside  $t_{bin}$ . That value favorably compares to the performances found in the literature [10] even without considering the factor of two enhancement brought by the multimode nature of the used pump source, that partially compensate for the lower quantum efficiency of the InGaAs detector we used, compared to superconducting nanowire single photon detectors.

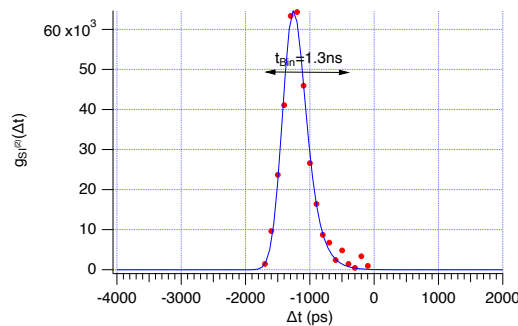


Figure 9: Plot of the histogram of  $g_{SI}^{(2)}$  around the coincidence peak, calculated with a value of  $t_{bin}=100ps$  corresponding to the temporal resolution of our counting and correlation system. The pump power for this measurement was 24mW, and acquisition time was 240s. The coincidence peak is fitted by an exponential-convoluted gaussian function (a pre-programmed function of the multipeak fitting procedure of IgorPro). It reaches a peak value around 65000 but is clearly larger than the temporal resolution value. The CAR value obtained with a value of  $t_{bin}=1300ps$  would lead to a reduced value of 24000 that seems to us more representative of the real performances of our source.

This analysis also shows that in the field of experimental characterization of CAR and  $g_{SI}^{(2)}$  the measurement procedures are still not completely normalized and that comparison of presented data from different sources with different structures must be realized carefully.

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