Supplementary Material for
Narrowband four-photon states from spontaneous four-wave mixing

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EVENT SEARCHING ALGORITHMS

The search for multiple coincidences over timestamp data from four detectors is a computationally resource intensive task. We employ the following strategy to simplify the search. We first identify pair coincidences at various relative delays for each pair from the following possible pairs of Stokes-anti-Stokes detections D1-D3, D1-D4, D2-D3 and D2-D4. The triplet coincidences are then identified based on the pair detections that share a photon arrival timestamp. Quadruplet events are identified from triplet detections that share a common timestamp with a pair detection.

For example, to identify triplet events involving D1, D3, and D4, we compare the timestamps of pairs between D1 and D3 detected at timestamps t_1 and t_3 with pairs between D1 and D4 detected at timestamps t_1' and t_4. Pair events that share a common timestamp in D1 i.e. $t_1 = t_1'$, are taken to form the triplet event (t_1, t_3, t_4). This information can be used to identify quadruplets detected across the four detectors. For this we compare pair events between D2 (at t_2) and D4 (at time t_4^*) with the previously identified triplet event with times t_1, t_3, t_4. When $t_4 = t_4^*$, the events are combined to form a quadruplet detection with the timestamp t_1, t_2, t_3, t_4. Thus, we can efficiently search for pair, triplet, and quadruplet events from timestamp data and plot the temporal distribution of these coincidences.

CORRELATED QUADRUPLETS VS UNCORRELATED DOUBLE PAIRS

We compare the four-photon component of the output in two different scenarios: when the output contains an aggregate of multiple independent SFWM events in the cloud vs when the output can be described as generated from a single coherent SFWM process. When a large number of independent SFWM processes take place any $2n$ photon state can be satisfactorily described as n independent pairs [1]. In this case, the probability of creating n pairs (P_n') within a certain time window, is described by a Poisson distribution of mean μ, $P_n' = e^{-\mu}\mu^n/n!$. For small μ, $P_4' \approx P_2'^2/2$. Here the four-photon states can be understood as a consequence of independent pairs that are coincidental in time. However, if the output is the consequence of a single coherent SFWM process, it is described by Eq. 1 in the main manuscript. At small interaction strengths ($\zeta \ll 1$) the probability of generating states with four photons (P_4) relates to the probability of producing pairs (P_2) as $P_4 = P_2^2$. Here the quadruplets are time-correlated. The probability of generating four-photon states in the latter scenario, is twice as much as in the former scenario.

The factor of two in the ratio between correlated four-photons and uncorrelated double pairs also manifests in a measurement of the third-order correlation between the modes as seen in Fig.3 in the manuscript. From Eq.9, when all delays are less than the coherence time (Δt) of the photons i.e., $\tau_{3s}, \tau_{34}, \tau_{4s} \lesssim \Delta t$, the third-order correlation increases several-fold. Using $g^{(2)}_{a,a}(0) = 2$ (thermal statistics in each of the two correlated modes), we get the limit $g^{(3)}_{a,a,s}(0,0,0) \leq 4g^{(2)}_{a,s}(0)$. The theoretical maximum possible value of $g^{(3)}_{a,a,s}(0,0,0) = 4g^{(2)}_{a,s}(0)$ is obtained when $g^{(2)}_{a,s}(0) \gg 1$, a condition true for highly non-classical pair sources. Thus, theoretically, the peak is expected to be 4 times the average in either the horizontal or vertical ridges, or 2 times the sum of horizontal and vertical ridges, when the output contains highly correlated four-photon states.

EXPANDED SECOND, THIRD AND FOURTH ORDER CORRELATION FUNCTIONS

Here we express the double, triple and quadruple coincidence rates in terms of the phase sensitive first-order cross-correlation,

$$C(\tau_{ij}) = \langle \hat{E}_i(t+\tau_{ij})\hat{E}_j(t)\rangle, \quad (1)$$

and the first-order autocorrelation

$$R(\tau_{ij}) = \langle \hat{E}_i^\dagger(t+\tau_{ij})\hat{E}_j(t)\rangle. \quad (2)$$

Here i and j can be s and/or a to represent the Stokes and/or anti-Stokes mode or $\{i,j\} \in \{1,2,3,4\}$ to represent one of the four detection channels. Note, $R(0)$ is the pair generation rate.

The second order intensity correlation between fields

$\hat{E}_i$ and $\hat{E}_j$ in modes i and j is

$$g^{(2)}_{ji}(\tau_{ji}) = \frac{\langle \hat{E}^\dagger_i(t_i)\hat{E}^\dagger_j(t_i+\tau_{ji})\hat{E}_j(t_i+\tau_{ji})\hat{E}_i(t_i)\rangle}{\langle \hat{E}^\dagger_j(t_i+\tau_{ji})\hat{E}_j(t_i+\tau_{ji})\rangle\langle \hat{E}^\dagger_i(t_i)\hat{E}_i(t_i)\rangle} . \tag{3}$$

When the state under consideration is the output of a parametric photon-pair production process such as SPDC or SFWM, the Gaussian moment factoring theorem can be applied to the intensity correlations in Eq.3 to give the following expressions for the normalized intensity cross-correlation [2]

$$g^{(2)}_{s,a}(\tau) = 1 + \frac{|C(\tau_{s,a})|^2}{|R(0)|^2}, \tag{4}$$

and the normalized intensity autocorrelation

$$g^{(2)}_{i,i}(\tau) = 1 + \frac{|R(\tau_{i,i})|^2}{|R(0)|^2}. \tag{5}$$

The triple-coincidence rate $G^{(3)}_{a,a,s}$ for an two anti-Stoke detections, one each at times t_3 and t_4 in D3 and D4 respectively, and a single Stokes detection at time t_s in either of D1 or D2

$$G^{(3)}_{a,a,s}(t_3,t_4,t_s) = \langle \hat{E}^\dagger_s(t_s)\hat{E}^\dagger_a(t_3)\hat{E}^\dagger_a(t_4)\hat{E}_a(t_4)\hat{E}_a(t_3)\hat{E}_s(t_s)\rangle, \tag{6}$$

can be similarly expanded and expressed in terms of relative delays to give [3, 4]

$$\begin{aligned} G^{(3)}_{a,a,s}(\tau_{3s},\tau_{4s},\tau_{34}) = & R(0)[R(0)^2 + |R(\tau_{34})|^2] \\ & + R(0)[|C(\tau_{3s})|^2 + |C(\tau_{4s})|^2] \\ & + 2\mathrm{Re}\{C(\tau_{3s})C^*(\tau_{4s})R(\tau_{34})\}. \end{aligned} \tag{7}$$

Thus the accidental triplet events are a sum of four contributions: accidental singles detected in each of the three channels, a correlated pair between the Stokes mode and D3 (D4) with an accidental in D4 (D3) and thermally bunched photons in the anti-Stokes (causing coincidences in D3 and D4) with an accidental single in the Stokes mode (D1 or D2). Using Eq. 4, and Eq. 5 this can be rewritten as

$$\begin{aligned} G^{(3)}_{a,a,s}(\tau_{3s},\tau_{4s},\tau_{34}) = & R(0)^3 g^{(2)}_{a,a}(\tau_{34}) + R(0)^3 g^{(2)}_{a,s}(\tau_{3s}) \\ & + R(0)^3 g^{(2)}_{a,s}(\tau_{4s}) - 2R(0)^3 \\ & + 2R(0)^3\sqrt{g^{(2)}_{a,s}(\tau_{3s})-1} \\ & \sqrt{g^{(2)}_{a,s}(\tau_{4s})-1} \times \sqrt{g^{(2)}_{a,a}(\tau_{34})-1}. \end{aligned} \tag{8}$$

The normalized third-order correlation is then

$$\begin{aligned} g^{(3)}_{a,a,s}(\tau_{3s},\tau_{4s},\tau_{34}) = & \frac{G^{(3)}_{a,a,s}(\tau_{3s},\tau_{4s},\tau_{34})}{R(0)^3} \\ = & g^{(2)}_{a,a}(\tau_{34}) + g^{(2)}_{a,s}(\tau_{3s}) \\ & + g^{(2)}_{a,s}(\tau_{4s}) - 2 \\ & + 2\sqrt{g^{(2)}_{a,s}(\tau_{3s})-1}\sqrt{g^{(2)}_{a,s}(\tau_{4s})-1} \\ & \times \sqrt{g^{(2)}_{a,a}(\tau_{34})-1}. \end{aligned} \tag{9}$$

Similarly, the quadruplet rate for detecting two Stokes photons at times t_1, and t_2 and two anti-Stokes photons at times t_3 and t_4,

$$G^{(4)}_{s,s,a,a}(t_1,t_2,t_3,t_4) = \langle \hat{E}^\dagger_a(t_4)\hat{E}^\dagger_a(t_3)\hat{E}^\dagger_s(t_2)\hat{E}^\dagger_s(t_1)\hat{E}_s(t_1)\hat{E}_s(t_2)\hat{E}_a(t_3)\hat{E}_a(t_4)\rangle. \tag{10}$$

On applying the Gaussian moment factoring theorem, we obtain the following expression in terms of relative delays.

$$\begin{aligned} G^{(4)}_{s,s,a,a}(\tau_{34},\tau_{24},\tau_{14},\tau_{23},\tau_{21},\tau_{13}) = & R(0)^4 \\ & + R(0)^2[|R(\tau_{43})|^2 + |R(\tau_{21})|^2] \\ & + R(0)^2[|C(\tau_{23})|^2 + |C(\tau_{24})|^2] \\ & + R(0)^2[|C(\tau_{13})|^2 + |C(\tau_{14})|^2] \\ & + 2R(0)\mathrm{Re}\{R(\tau_{34})C^*(\tau_{24})C(\tau_{23})\} \\ & + 2R(0)\mathrm{Re}\{R(\tau_{34})C^*(\tau_{14})C(\tau_{13})\} \\ & + 2R(0)\mathrm{Re}\{R(\tau_{21})C^*(\tau_{14})C(\tau_{24})\} \\ & + 2R(0)\mathrm{Re}\{R(\tau_{21})C^*(\tau_{13})C(\tau_{23})\} \\ & + |R(\tau_{43})|^2|R(\tau_{21})|^2 \\ & + |C(\tau_{13})|^2|C(\tau_{24})|^2 + |C(\tau_{14})|^2|C(\tau_{23})|^2 \\ & + 2\mathrm{Re}\{C^*(\tau_{24})C^*(\tau_{13})C(\tau_{14})C(\tau_{23})\}] \\ & + 2\mathrm{Re}\{R(\tau_{34})R(\tau_{12})C^*(\tau_{24})C(\tau_{13})\} \\ & + 2\mathrm{Re}\{R(\tau_{34})R(\tau_{21})C(\tau_{23})C^*(\tau_{14})\} \end{aligned} \tag{11}$$

From the 17 terms in Eq 11, all terms other than the last three are contributions due to accidentals. Terms 2-7, are due to two accidentals combined either with a correlated Stokes-anti-Stokes pair or with thermally bunched photons in either of the Stokes or anti-Stokes modes. Terms 8-11 are caused by an accidental combined with a correlated triplet in three detectors. Term 12 is from bunching in both the Stokes and anti-Stokes modes. Terms 13 and 14 are correlated pairs from separate SFWM events contributing to four-fold coincidences. Terms 15, 16 and 17 are due to correlated double-pairs from the same SFWM event.

ACCIDENTAL CORRECTION PROCEDURE

Correction must be performed to eliminate contributions from accidental coincidences between uncorrelated events from different channels. This can be visualized as the relative probability of independent events falling within the same coincidence time window t_c (i.e. random chance). For coincidences between two detectors, the accidental rate is $t_c R_i R_j$ for singles rates R_i and R_j in detectors Di and Dj respectively. Excess coincidence events after correction can then only be attributed to actual correlations between channels: in the case of two detectors Di and Dj with an observed pair rate of R_{ij}, the correlated pair rate c_{ij} is given by

$$c_{ij} = R_{ij} - t_c R_i R_j. \tag{12}$$

The total correlated pair rate is $c_p = \sum_{i,j} c_{ij}$ for $\{i,j\} \in \{\{1,3\},\{1,4\},\{2,3\},\{2,4\}\}$.

These correlated pairs (and separately, accidentals) can be modeled as a separate stream of events that factor into the calculation of higher-order accidentals, e.g. 3-fold accidentals between channels i, j and k occur due to accidental coincidences across the individual channels $\{R_i, R_j, R_k\}$, as well as pairs with the remaining channel $\{c_{ij}, R_k\}$, $\{c_{ik}, R_j\}$ and $\{c_{jk}, R_i\}$. For instance, in Fig. 3 in the manuscript, these correspond to the general background horizontal, vertical ridges and diagonal ridges when $\{i,j,k\} = \{s,3,4\}$.

The correlated triplet rate is thus

$$c_{ijk} = R_{ijk} - t_c \left(c_{ij} R_k + c_{jk} R_i + c_{ik} R_j\right) - t_c^2 R_i R_j R_k \tag{13}$$

given an observed triplet rate of R_{ijk}.

It can be seen that each of the individual terms contributing to the n-fold coincidences correspond to a possible partitioning of the set of all channels, with the total number of partitions given by Bell's number B_n (i.e. $B_2 = 2$, $B_3 = 5$, $B_4 = 15$). We write out explicitly the exhaustive 14-term correction performed for 4-fold coincidences,

$$\begin{aligned} c_{1234} = R_{1234} \\ & - t_c \left(c_{12}c_{34} + c_{13}c_{24} + c_{14}c_{23}\right) \\ & - t_c \left(c_{123}R_4 + c_{124}R_3 + c_{134}R_2 + c_{234}R_1\right) \\ & - t_c^2 \left(c_{12}R_3R_4 + c_{13}R_2R_4 + c_{14}R_2R_3\right) \\ & - t_c^2 \left(c_{23}R_1R_4 + c_{24}R_1R_3 + c_{34}R_1R_2\right) \\ & - t_c^3 R_1R_2R_3R_4. \end{aligned} \tag{14}$$

We also make a small note that this correction slightly overestimates the actual accidental rate [5] due to the n-fold coincidence calculation method containing an implicit ordering of events that introduces excess accidentals.

In the experiment, this overcompensation is minimized by using a small 20 ns coincidence window. At fixed pump and coupling powers of 800 μW and 10 mW respectively, and $\Delta_p = 40$ MHz, the mean value of singles rates for Stokes photons is $R_s = (1.04 \pm 0.07) \times 10^6$ cps and anti-Stokes photons $R_a = (1.10 \pm 0.06) \times 10^6$ cps. Performing accidental subtraction as outlined above we obtained a correlated-pair detection rate $c_p = (4.8 \pm 0.3) \times 10^4$ cps.

As we can see from Eq.13, the accidental corrected rate depends on the combination of detectors chosen. Thus, we report detector-specific accidental-corrected triplet rates. The corrected rate of detected photon triplets consisting of one Stokes photon and one anti-Stokes photon each in D3 and D4 is $c_{134} + c_{234} = (251 \pm 10)$ cps, while the rate for triplets consisting of a Stokes photon each in D1 and D2 and one anti-Stokes photon is $c_{123} + c_{124} = (246 \pm 7)$ cps. The correlated photon quadruplet rate after accidental-subtraction, for a detection in each of the four detectors, is found to be $c_q = (2.9 \pm 0.4)$ cps.

We infer the generation rates of pairs g_p and double-pairs g_q from detected accidental-corrected rates of pairs, triplets and quadruplets by factoring in the losses as described in . Most of the accidentals are dominated by coincidences between uncorrelated souble pairs (5.5 cps), followed by coincidences from a pair and two singles (5.6 cps) and coincidences from a triplet combined with a single (4.8 cps).

Code for the accidental correction as well as the corresponding datasets can be found in...

CHANNEL LOSSES

We characterize the losses in each channel to estimate the rate of correlated pairs and double-pairs directly generated from the SFWM process. The total transmission and detection probability in each channel $k \in \{1,2,3,4\}$, which includes transmission of the collection and filtering setup, splitting efficiency of the fiber based 50:50 beamsplitter and quantum efficiency of the detector in the respective channel is denoted by η_k. The optical losses in each channel are determined by measuring the transmission of a laser beam (at the target wavelength) from outside the vacuum chamber to just before the detector in each channel. We measure transmissions of $\approx 11.5\%$ each for channels 1 and 2 (with detectors D1 and D2 respectively) and 12.5% each for channels 3 and 4 (corresponding to D3 and D4 respectively), which include the contributions from the filter-Etalon and fiber beamsplitter. Including the quantum efficiencies of avalanche photodiodes used as single photon detectors in each channel (about 60-70% per detector), the measured efficiencies (η'_k) are 0.078, 0.083, 0.080, and 0.067 for channels 1,2,3 and 4 respectively. These values provide an estimate of the upper bound for effective efficiencies, as they do not account for absorption in the atomic ensemble or spatial mode mismatch between the photons and the collection optics.

Since we expect additional losses that are frequency specific to the Stokes and anti-Stokes modes, we define $\eta_i = \eta_s \eta_i'$ for $i \in \{1,2\}$ and $\eta_j = \eta_a \eta_j'$ for $j \in \{3,4\}$. We then use equations 16 and 15 to estimate η_s and η_a. We infer additional losses that are $1-\eta_s = 19\%$ for channels in the Stokes arm and $1-\eta_a = 8\%$ for the anti-Stokes channels, which we attribute to a combination of above mentioned factors. Taking into account these additional losses, the total efficiencies are $\eta_1 = 0.022$ and $\eta_2 = 0.023$ for channels 1 and 2 pertaining to the Stokes modes, and the total efficiencies are $\eta_3 = 0.025$ and $\eta_4 = 0.021$ for channels 3 and 4 of the anti-Stokes mode.

GENERATION RATES FROM DETECTION RATES

We infer the generation rates of pairs g_p and correlated double-pairs g_q from the detected accidental-corrected rates of pairs, triplets and quadruplets by factoring in the channel losses as follows. c_q, the accidental-corrected quadruplet rate, is solely contributed to by generated correlated double-pairs. There are four possible combinations by which each of the two Stokes photons reach D1 and D2 and each of the two anti-Stokes photons reach D3 and D4. This gives,

$$c_q = 4 g_q \eta_1 \eta_2 \eta_3 \eta_4, \tag{15}$$

Similarly, double-pair generations are the sole contributors to accidental-corrected triplet coincidences. A triplet between detectors D1, D3 and D4 occurs from two possible combinations by which the two anti-Stokes photons reach one of D3 and D4 each (leading to the factor $2\eta_3\eta_4$) combined with the probability that at least one of the two Stokes photons reaches D1 (leading to the factor $1-(1-\eta_1)^2$ where $(1-\eta_1)^2$ is the probability that neither of the two Stokes photons reaches D1). Applying this to all combinations of triplet detections,

$$\begin{aligned} c_{123} &= 2g_q\eta_1\eta_2(2-\eta_3)\eta_3 \\ c_{124} &= 2g_q\eta_1\eta_2(2-\eta_4)\eta_4 \\ c_{134} &= 2g_q\eta_3\eta_4(2-\eta_1)\eta_1 \\ c_{234} &= 2g_q\eta_3\eta_4(2-\eta_2)\eta_2. \end{aligned} \tag{16}$$

Here c_{ijk} is the rate of correlated triplets between detectors i, j and k. We use this to estimate mean values of g_q.

Since we have non-number-resolving detectors, both pairs and correlated double-pairs from the SFWM process contribute to accidental-corrected pair coincidences. A coincidence between D1 and D3 can be caused by a generated pair where the Stokes photon is detected in D1 and the anti-Stokes is detected in D3 ($\eta_1\eta_3$) or the probability that at least one of two Stokes and two anti-Stokes photons from a double-pair reach D1 and D3 respectively $((1-(1-\eta_1)^2)(1-(1-\eta_3)^2) = \eta_1\eta_3(2-\eta_1)(2-\eta_3))$. This gives,

$$\begin{aligned} c_{13} &= \eta_1\eta_3(g_p + g_q(2-\eta_1)(2-\eta_3)) \\ c_{14} &= \eta_1\eta_4(g_p + g_q(2-\eta_1)(2-\eta_4)) \\ c_{23} &= \eta_2\eta_3(g_p + g_q(2-\eta_2)(2-\eta_3)) \\ c_{24} &= \eta_2\eta_4(g_p + g_q(2-\eta_2)(2-\eta_4)) \end{aligned} \tag{17}$$

where c_{ij} is the accidental-corrected pair rate between channels i and j.

We use use accidental-corrected pair, triplet and quadruplet rates to obtain values for g_p and g_q, reported in the manuscript

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