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LETTERS

Direct generation of photon triplets using cascaded photon-pair sources

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Non-classical states of light, such as entangled photon pairs and number states, are essential for fundamental tests of quantum mechanics and optical quantum technologies. The most widespread technique for creating these quantum resources is spontaneous parametric down-conversion of laser light into photon pairs[1]. Conservation of energy and momentum in this process, known as phase-matching, gives rise to strong correlations that are used to produce two-photon entanglement in various degrees of freedom[2–9]. It has been a longstanding goal in quantum optics to realize a source that can produce analogous correlations in photon triplets, but of the many approaches considered, none has been technically feasible[10–17]. Here we report the observation of photon triplets generated by cascaded down-conversion. Each triplet originates from a single pump photon, and therefore quantum correlations will extend over all three photons[18] in a way not achievable with independently created photon pairs[19]. Our photon-triplet source will allow experimental interrogation of novel quantum correlations[20], the generation of tripartite entanglement[12,21] without post-selection and the generation of heralded entangled photon pairs suitable for linear optical quantum computing[22]. Two of the triplet photons have a wavelength matched for optimal transmission in optical fibres, suitable for three-party quantum communication[23]. Furthermore, our results open interesting regimes of non-linear optics, as we observe spontaneous down-conversion pumped by single photons, an interaction also highly relevant to optical quantum computing.

Given the potential for fundamental and applied quantum sciences, several physical systems have been proposed for the direct generation of photon triplets. These include four-level atomic cascades and higher-order optical nonlinearities[10], tri-excitons in quantum dots[11], combinations of second-order nonlinearities[13] and high-energy electron–positron collisions[14]. Extremely low interaction strengths and collection efficiencies have rendered these proposals unfeasible. Recent experiments have observed and studied third-order[15,16] and cascaded second-order nonlinear[17] parametric processes seeded by strong lasers. However, such seeding only increases stimulated emission, which masks the production of tripartite quantum correlations and cannot lead to three-photon entanglement.

Production of photon triplets by cascaded spontaneous parametric down-conversion (C-SPDC) was first proposed 20 years ago[12], yet never experimentally realized. The basic idea is shown in Fig. 1a. A primary down-conversion source is pumped by a laser to create a photon pair. One of the photons from this pair drives a secondary down-conversion process, generating a second pair and, hence, a photon triplet. Because the photon triplet originates from a single pump photon, the created photons have strong temporal correlations[24] and their energies and momenta sum to those of the original photon.

The C-SPDC process can be described using a simplified quantum optical model. The interaction Hamiltonian for the primary source can be written as $\mathcal{H}_1 = \lambda_1 \alpha (a_0^\dagger a_1^\dagger + \text{h.c.})$ (h.c., Hermitian conjugate), with the pump laser treated as a classical field with amplitude α and the photon creation operators of the two output modes denoted by $a_0^\dagger$ and $a_1^\dagger$, respectively. The coupling strength between the interacting fields is expressed by the parameter λ_1, which includes the nonlinear response of the material and governs the expected conversion rate of pump photons. For the second down-conversion, the pump field is a single photon and must be treated quantum mechanically in the interaction Hamiltonian, $\mathcal{H}_2 = \lambda_2 (a_0 a_2^\dagger a_3^\dagger + \text{h.c.})$, with output modes 2 and 3. The evolution operator of the system is $U = U_2 U_1 = \exp(-i\mathcal{H}_2)\exp(-i\mathcal{H}_1)$, and can be approximated by expanding each term to first order. Applying U to the initial vacuum state and ignoring the vacuum contribution for the final state results in

$$
\begin{aligned}
|\Phi\rangle &= U|0_0, 0_1, 0_2, 0_3\rangle \\
&\approx -i\lambda_1\alpha|1_0, 1_1, 0_2, 0_3\rangle - \lambda_1\lambda_2\alpha|0_0, 1_1, 1_2, 1_3\rangle
\end{aligned}
\tag{1}
$$

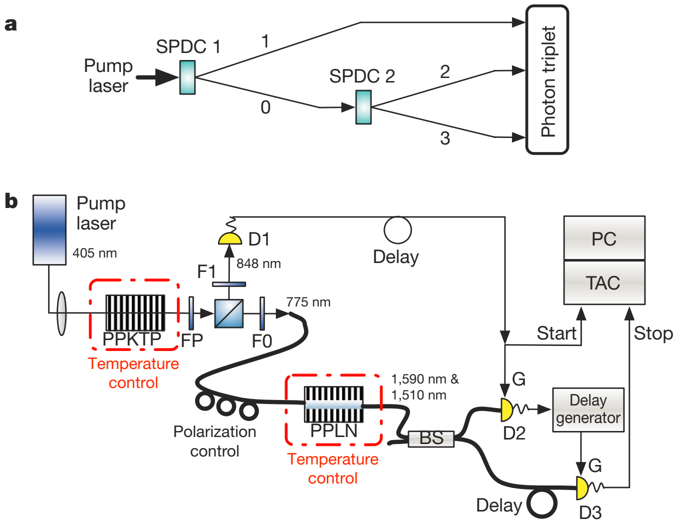

Figure 1 | Schematic of photon-triplet generation and experimental set-up. **a**, A down-conversion source (SPDC 1) produces a pair of photons in spatial modes 0 and 1, where the photon in mode 0 creates another photon pair in the second source (SPDC 2) in modes 2 and 3, generating a photon triplet. **b**, The primary source, pumped by a 405-nm laser, produces photon pairs at 775 nm and 848 nm. The 848-nm photon is directly detected by a silicon avalanche photodiode (D1), and the 775-nm photon serves as input to the secondary source, creating a photon pair at 1,510 nm and 1,590 nm that is detected by two InGaAs avalanche photodiodes (D2 and D3). A detection event at D3 represents a measured photon triplet. BS, beam splitter; F0, F1, band-pass filters; FP, long-pass filter; G, gate; TAC, time acquisition card; PC, computer.

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where the subscripts label the spatial modes. The first term describes the pair creation process in the first crystal and the second represents the desired three-photon state, $|0, 1, 1, 1\rangle$, where the amplitude scales as the product of the two coupling strengths, λ_1 and λ_2, of the down-converters. Equation (1) predicts that the rate of triplet production from C-SPDC should be linear in the intensity of the pump laser.

The conversion efficiencies in SPDC are typically very low. In optical nonlinear materials such as β-barium borate, for example, they reach about 10^{-11} per pump photon[25]. Major advances in nonlinear optics, such as quasi-phase-matching of optical materials, have recently made it possible to access the inherent higher nonlinearities of materials such as periodically poled lithium niobate (PPLN) and periodically poled potassium titanyl phosphate (PPKTP). The down-conversion efficiencies demonstrated in these materials can reach up to 10^{-9} in bulk[26]. The introduction of optical waveguides in photon-pair sources[27] has further increased conversion efficiencies to 10^{-6}, making the observation of C-SPDC possible.

Figure 1b depicts the experimental set-up (see Methods for more details). The primary source generated photon pairs in a PPKTP crystal quasi-phase-matched for collinear SPDC of 405 nm → 775 nm + 848 nm. The 775-nm photons were used to pump the secondary source, which consisted of a PPLN waveguide quasi-phase-matched for 775 nm → 1,510 nm + 1,590 nm. The photon triplets were measured using a chained series of three photon counters (D1, D2 and D3) based on avalanche photodiodes. The detection of a 848-nm photon at D1, which occurred with a frequency of about 1 MHz, opened a 20-ns gate at D2, which in turn gated D3 for 1.5 ns. The actual gate rate of D2 was reduced to 870 kHz, owing to saturation. Because D3 was only armed if both D1 and D2 had fired, an event at D3 constituted the detection of a photon triplet. The temporal signatures of these triple coincidences were recorded in histograms with a fast time acquisition card, where the detection signal at D1 served as the start trigger and the detection signal at D3 served as the stop trigger. Data were recorded for a total of 20 h and analysed as a histogram of the time interval between detections at D3 and D1, $\Delta\tau_{D3-D1}$.

A typical data set, shown as a histogram in Fig. 2a, displays a peak 8 s.d. above the background noise. This is a clear signature of C-SPDC photon triplets. The 1.2-ns temporal width of the observed photon-triplet peak is dominated by detector jitter. Integration over the three central time bins yields a raw triplet rate of 124 ± 11 events in 20 h. The observed background in the histogram is caused predominantly by triple events between a genuine detection at D1 and dark counts at D2 and D3 (see Methods), and was estimated from the displayed data to be 10.2 ± 0.9 per bin in 20 h. The detected rate of triplets, exclusively produced by the C-SPDC process, was 4.7 ± 0.6 counts per hour. We modelled the process under the assumption that the down-conversion efficiency per photon in the secondary source was independent of the pump intensity (Supplementary Information). Using the conversion efficiencies obtained from independent characterizations of both sources at milliwatt pump power, and optical parameters from other relevant components of our set-up, our model predicts a triplet rate of 5.6 ± 1.1 counts per hour, which is in very good agreement with the measured value.

It is expected that C-SPDC photon triplets should exhibit strict time correlations[24]. We investigated this property by introducing three different delays between D2 and D3 (−0.5, 0 and 0.5 ns) and measuring the histograms. The data in Fig. 2b show a significant reduction of the peak in the histograms with additional delays, verifying the strong temporal correlations of the created triplets.

It is conceivable that other physical processes, such as the avalanche photodiode breakdown flash from D1[28], electronic cross-talk or double-pair emission from the primary source, might give rise to correlated triple-detection events with similar features to the ones we have observed. We can rule out these alternatives by testing the expected dependence of the C-SPDC signal on temperature and input wavelength of the secondary down-conversion. As shown in Fig. 3a, for a given input wavelength into the PPLN crystal, phase-matching imposes a minimum temperature below which down-conversion cannot occur. The triple-coincidence peak, in Fig. 3b, indeed disappears when the PPLN temperature is lowered from 60 °C (setting A) to 50 °C (setting B) while keeping the input wavelength fixed at 776.0 nm. The triple-photon signal is then recovered at this temperature by lowering the input wavelength to 775.4 nm (setting C). These measurements, together with

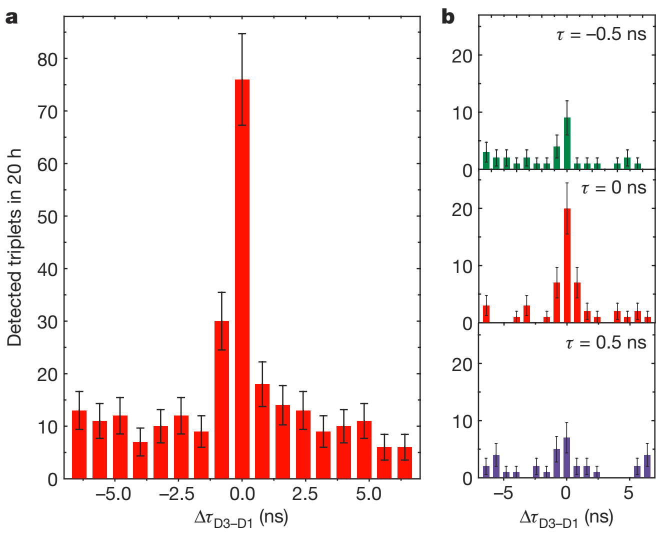

Figure 2 | Triple-coincidence histograms. **a**, Measured triple coincidences obtained in 20 h. Each bin corresponds to a 0.8-ns time interval between events at D3 and D1 ($\Delta\tau_{D3-D1}$). The sharp peak indicates a strong temporal correlation between all three detection events, as expected of the C-SPDC process. **b**, Triple-coincidence histograms with varying delays of $\tau = 0$ and ±0.5 ns between D2 and D3, resulting in a decrease of the coincidence peak. The absolute rate reduction for $\tau = 0$ results from a different setting on the InGaAs detectors for this measurement series. Error bars, 1 s.d.

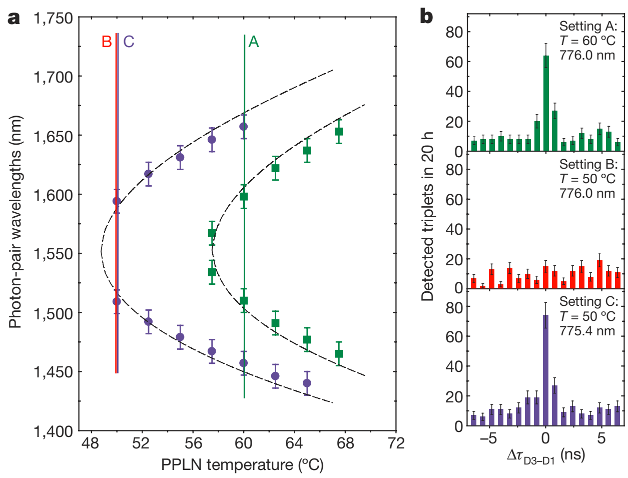

Figure 3 | Phase-matching and triple-coincidence dependence on crystal temperatures. **a**, Central wavelengths of the pair of photons produced by the secondary source as a function of the PPLN temperature for input wavelengths of 775.4 nm (circles) and 776.0 nm (squares). The dashed lines show the theoretical phase-matching curves with the poling period as the only fit parameter. Triple coincidences were measured for different settings of the PPLN temperature and the input pump photon wavelength. The PPLN temperature was 60 °C for setting A and 50 °C for settings B and C; the input photon wavelength was 776.0 nm for settings A and B and 775.4 nm for setting C. **b**, Measured triple coincidence histograms over 20 h for each measurement setting. For A and C, the PPLN temperatures lie on the respective phase-matching curves and a triple-coincidence peak is observed. For B, the temperature is outside the 776.0-nm phase-matching curve and no peak is present. Wavelength changes in the input photons, needed for the measurements shown in Fig. 3b, were achieved by altering the temperature of the PPKTP crystal (43.6 °C for settings A and B, 40.8 °C for C). Error bars, 1 s.d.

the strong agreement between the observed and predicted triplet rates, provide conclusive proof that we have indeed observed spontaneously produced photon triplets.

In the near future, we expect to increase the photon-triplet rate by at least one order of magnitude using an improved time acquisition system, a dichroic beam splitter for separating the photons created in the secondary source and by matching the down-conversion bandwidth of the initial pair to the PPLN crystal. The direct generation of the triplet guarantees strong energy–time correlations, allowing the creation of entangled, or hyper-entangled[29], triplets and realizations of tripartite states such as the Greenberger–Horne–Zeilinger (GHZ) state[30] and the W state[31] without elaborate and probabilistic post-selection schemes. For example, time-bin entangled[6] GHZ states could be produced by pumping our triplet source with a pulsed pump laser in a coherent superposition of two time slots. The entanglement could then be detected using three standard unbalanced interferometers. As a further example, W states could be made by using an entangling source as the primary down-converter, producing a Bell state $(|V_0V_1\rangle + |H_0H_1\rangle)/\sqrt{2}$, where $|V\rangle$ and $|H\rangle$ denote the photon polarization states in their respective modes. The secondary source would consist of two down-converters where $|V_0\rangle$ is converted to $|H_2H_3\rangle$ and $|H_0\rangle$ is converted to $(|H_2V_3\rangle + |V_2H_3\rangle)/\sqrt{2}$, into the same pair of modes. The relative amplitudes could then be balanced by tuning the conversion efficiencies. Polarization-entangled GHZ states could be made by modifying the W-state scheme such that the secondary source converts $|V_0\rangle$ to $|V_2V_3\rangle$ and $|H_0\rangle$ to $|H_2H_3\rangle$. An interesting application of such a GHZ source could be to indicate the presence of an entangled photon pair in modes 1 and 2 by detecting the secondary down-converted photon in mode 3. This has proven very difficult to achieve otherwise. Our results also confirm that the SPDC efficiency is independent of pump power down to the single-photon level (Supplementary Information), allowing new tests of nonlinear optics in the quantum regime.

METHODS SUMMARY

Experimental set-up. The primary source, shown in Fig. 1b, consisted of a 25-mm-long, temperature-stabilized PPKTP crystal and was pumped with a power of 2.4 mW from a 405-nm continuous-wave diode laser. The type-II SPDC in the PPKTP generated orthogonally polarized photons at 775 nm and 848 nm that were separated by a polarizing beam splitter and coupled into single-mode fibres. A long-pass filter (FP) was used to block the strong 405-nm pump, band-pass filters (12-nm bandwidth) with respective central wavelengths of 780 nm (F0) and 840 nm (F1) were placed before the fibre couplers to further reduce background. The 775-nm photon, after passing an in-fibre polarization controller, served as input to the secondary source, a 30-mm temperature-stabilized PPLN waveguide crystal with fibre pigtails attached to both ends for type-I SPDC. The photon pair at 1,510 nm and 1,590 nm was separated using a 50:50 fibre beam splitter (BS). The secondary source was operated without filters, as the input power during C-SPDC measurements was low enough ($\sim 10^6$ input photons per second) not to cause additional detection events in the InGaAs detectors. The gate (G) and photon arrivals at these detectors were synchronized by an internal delay generator at D2 and an external delay generator between D2 and D3. Detection efficiencies at the InGaAs detectors D2 and D3 were set to 20% and 10%, respectively. Trigger events from D1 and detection events from D3 were recorded using a time acquisition card (TAC) with a timing resolution of 103 ps, and analysed on a computer (PC).

Dark count rate. The total background during the 20-h runs, seen in Fig. 2a, was measured to be 268 ± 16 events over the whole 20-ns gate. This number is in very good agreement with the expected noise count of 254 ± 5 triple events as calculated from the individual dark count probabilities per gate of D2 (1.8×10^{-3}) and D3 (4.5×10^{-6}), the trigger rate and the efficiency of the time acquisition card.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Author Contributions H.H. and D.R.H. performed the experiment and analysed the data; A.F. and S.R. participated in the design of the experiment; K.J.R. and T.J. contributed to the design and realization of the experiment; and all authors co-wrote the paper.

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