Title: Generation of multi-photon entangled quantum states via integrated frequency combs

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Abstract:
Complex optical photon states with entanglement shared among several modes are critical in improving our fundamental understanding of quantum mechanics, and find application in quantum information processing as well as in quantum imaging and microscopy. We demonstrate that optical integrated Kerr frequency combs can be used to generate several bi- as well as multi-photon entangled qubits, which can find direct applications in quantum communication and computation. Our method is compatible with contemporary fiber and quantum memory infrastructures, as well as chip-scale semiconductor technology, enabling compact, low-cost, and scalable implementations. The exploitation of integrated Kerr frequency combs, and their ability to generate multiple, customizable, complex quantum states, can provide a scalable and practical platform for quantum technologies.

Main Text:
Multi-entangled states of light hold answers to fundamental questions in quantum physics and are the cornerstone of a large variety of applications including quantum communications (*1*), computation (*2–4*), as well as sensing and imaging with a resolution beyond the classical limit (*5*). Thus, the controllable realization of multiple quantum states in a compact platform would

provide for a practical and powerful implementation of quantum technologies. While frequency comb applications have been mostly classical, their unique architecture, based on multiple interacting modes as well as the phase characteristics of the underlying nonlinear processes, has the potential to offer new and powerful ways to achieve the generation of multiple, customizable, complex states of non-classical light. Indeed, the quantum properties of frequency combs have recently begun to be investigated, revealing their potential for the generation of large quantum states (*6–8*). However, the continuous-variable non-classical states (i.e. squeezed vacuum) that have been demonstrated with this approach have not yet achieved the quality (i.e. amount of squeezing) required for optical quantum computation (*9*). For the generation of single photons, continuous-, as well as discrete-variable quantum states (i.e. qubits), a large variety of second- and third-order nonlinear sources, optical fibers, gases, as well as single quantum emitters, have been exploited (*10*, *11*). Recent progress has focused on transferring both classical frequency combs (*12*), as well as quantum sources (*13*) to integrated optical platforms. Such integrated approaches provide the advantages of compact, scalable, mass-producible, and low-cost devices (*14*). Demonstrated integrated devices include sources of heralded single photons (*15–17*), entangled photon pairs (*18*) as well as the implementation of quantum algorithms (*19*, *20*). Here we show the parallel generation of bi- and multi-photon entangled states in a compact integrated quantum frequency comb source.

Our quantum frequency comb is generated in a CMOS (Complementary Metal–Oxide–Semiconductor) compatible high refractive index glass in a four-port microring resonator architecture (details on device fabrication and characteristics are presented in (*21*)). The weak and anomalous dispersion of our device enables broadband phase matching for spontaneous Four-Wave Mixing (SFWM), generating a broad frequency comb of photons emitted at the cavity resonances. The ring resonator's high Q-factor of around 240,000 (803 MHz linewidth, and 200 GHz free spectral range, see Fig. S1) allows for high field-enhancement and low-power operation, while simultaneously enabling the direct generation of bright and narrow-bandwidth photons, without the need for spectral filtering of the photons (*22*, *23*). The device was pumped with a 16.8 MHz repetition rate passive mode-locked fiber laser, spectrally filtered to excite a single ring resonance at 1556.2 nm (192.65 THz) with a pulse duration of 570 ps coupled into the resonator. We note that the pulses are directly filtered by the resonator, resulting in a perfect match between the spectral bandwidth of the pump pulses and the excited resonator mode. This in turn leads to the generation of pure single-mode photons per resonance (*13*), confirmed by single photon auto-correlation measurements (Fig. S2; see (*21*) for further details).

Using a high-resolution tunable C-band wavelength filter as well as a grating-based spectrum analyser, the single photon spectrum was characterized at the output of the resonator (*21*). Figure 1 shows the measured single photon count rate and the calculated photon pair production rate per pulse as a function of wavelength. It can be clearly seen that a very broad frequency comb of photons is emitted, covering the full S, C, and L International Telecommunication Union (ITU) bands (wavelengths ranging from 1470 to 1620 nm). The SFWM process generates a spectrum symmetric in frequency, while the spectral asymmetry in the measured photon counts can be explained by Raman scattering, which could be further reduced by cooling the chip (*24*). Due to the broad phase-matching condition, achieved through the close-to-zero waveguide dispersion, the emitted comb exhibits a flat and broadband spectrum with uniform pair production rates, ranging from 0.02 to 0.04 pairs per pulse, over the full measured comb.

To demonstrate an entangled quantum frequency comb, we chose time-bin entanglement, which among several intrinsic advantages, is particularly suitable for information processing and transmission (*25*) because of its robustness (e.g. with respect to polarization fluctuations), and thus can be preserved even over long propagation distances in standard fiber networks (*26*). Starting from the single-mode photon pairs, we generated time-bin entangled qubits by passing the pulsed pump laser through a stabilized unbalanced fiber interferometer with a 11.4 ns delay – larger than the pulse duration of the laser – generating double pulses of equal power with a defined relative phase difference. The temporal separation of the two pulses can be arbitrarily chosen as long as it is larger than the temporal duration of the single photons, thus offering significant flexibility. The double pulses were then coupled into the integrated microring resonator (Fig. 2), where an average pump power of 0.6 mW was chosen in such a way that the probability of creating a photon pair from both pulses simultaneously was sufficiently low to be negligible. This pump configuration transforms the originally single-mode photon pairs into entangled states, where the photons are in a superposition of two temporal modes. In particular, the entangled state $|\Psi_{time-bin}\rangle = \frac{1}{\sqrt{2}}(|\mathrm{S}_s, \mathrm{S}_i\rangle + |\mathrm{L}_s, \mathrm{L}_i\rangle)$ is generated, where the signal (s) and idler (i) photons are in a quantum superposition of the short (S) and long (L) time-bin (see (*21*) for more details). Most importantly, these entangled qubits are generated over all the microring resonances, thus leading to a quantum frequency comb of time-bin entangled photon pairs.

To characterize the degree of entanglement, the generated signal and idler photons were each passed individually through a different fiber interferometer with an imbalance identical to that used for the pump laser (Fig. 2). This setup allowed the measurement of the quantum interference between signal and idler photons. For the resonances closest to the excitation frequency a photon coincidence rate of 340 Hz was measured, leading to an estimated pair production rate of 302 kHz per channel (0.018 pairs per double pulse), accounting for system and detection losses of 14.75 dB (see (*21*) for more details). We selected 5 different frequency channel pairs within the ITU C band (marked in Fig. 1), and recorded quantum interference with raw visibilities above $0.824 > 1/\sqrt{2} \approx 0.71$ (top inset of Fig. 1, and Fig. 3) thus confirming entanglement through the violation of the Clauser-Horne-Shimony-Holt (Bell-like) inequality (*27*). After subtracting the measured background (also shown in Fig. 3), the visibility is found to be above 93.2% on all channel pairs (top inset in Fig. 1).

In addition to confirming time-bin entanglement, the quantum interference also reveals the phase-characteristic of the nonlinear generation process. Indeed, this phase dependency has been well described in theory (*28*) and has been exploited, for example, in optical squeezing (*29*). Here we show that this phase dependency is also manifested at the single photon level with a clear difference between second- and third-order nonlinear interactions. When Spontaneous Parametric Down-Conversion (SPDC) in second-order nonlinear media is used to generate the entangled photon pairs, quantum interference is expected to be proportional to $1 - V cos(\alpha + \beta - \varphi)$ (*26*), where V is the fringe visibility, φ is the pump interferometer phase, and α and β are the phases for the signal and idler interferometers, respectively. In contrast, for photons generated through SFWM (as we do in this work), quantum interference is expected to be of the form $1 - V cos(\alpha + \beta - 2\varphi)$ for degenerate SFWM, or $1 - V cos(\alpha + \beta - \varphi_1 - \varphi_2)$ for non-degenerate SFWM, where $\varphi_{1,2}$ are the phases of the two pump fields (*21*). As shown in Fig. 3, we confirm this important difference in phase dependency between SPDC and SFWM through five separate quantum interference measurements, where the phases of the interferometers are either tuned separately or simultaneously in a symmetric and anti-symmetric way. The expected behaviour for SPDC and SFWM is plotted in Figs. 3A-E with dashed and solid lines, respectively. The measurements clearly confirm the difference between the two processes, which can be explained through the additional photon involved in the SFWM process. In the generation of entangled photon pairs, the phase of the excitation photon(s) can be used to adjust the quantum state. If a single photon is involved (as in second-order processes), only the absolute phase of this photon can be used as a control parameter. In third-order nonlinear processes, however, two photons generate the quantum state, enabling an additional control parameter (i.e. the relative phase between the two photons, in turn much easier to control). While here only a single excitation field was used to demonstrate the effect, exploiting two distinct non-degenerate pump fields could lead to an additional degree of freedom for all-optical reconfigurable quantum state generation.

The unique multi-mode characteristic of the frequency comb architecture presented here can be extended to create multi-photon entangled quantum states using the same platform. By selecting two different signal-idler pairs we can generate two-photon qubit states given by $|\Psi_1\rangle = \frac{1}{\sqrt{2}}\left(|\mathrm{S}_{s1}, \mathrm{S}_{i1}\rangle + e^{i2\varphi}|\mathrm{L}_{s1}, \mathrm{L}_{i1}\rangle\right)$ and $|\Psi_2\rangle = \frac{1}{\sqrt{2}}\left(|\mathrm{S}_{s2}, \mathrm{S}_{i2}\rangle + e^{i2\varphi}|\mathrm{L}_{s2}, \mathrm{L}_{i2}\rangle\right)$. By post-selecting four-photon events with one photon on each frequency channel, these two states are multiplied, resulting in a four-photon time-bin entangled state, given by $|\Psi_{4-photon}\rangle = |\Psi_1\rangle \otimes |\Psi_2\rangle = \frac{1}{2}\left(|\mathrm{S}_{s1}, \mathrm{S}_{i1}, \mathrm{S}_{s2}, S_{i2}\rangle + e^{i2\varphi}|S_{s1}, \mathrm{S}_{i1}, \mathrm{L}_{s2}, \mathrm{L}_{i2}\rangle + e^{i2\varphi}|\mathrm{L}_{s1}, \mathrm{L}_{i1}, \mathrm{S}_{s2}, \mathrm{S}_{i2}\rangle + e^{i4\varphi}|L_{s1}, \mathrm{L}_{i1}, \mathrm{L}_{s2}, \mathrm{L}_{i2}\rangle\right)$. It is worth mentioning that for the generation of a four-photon state, the coherence length of both generated photon pairs has to be the same and matched to the excitation field's coherence time. This requirement is intrinsically fulfilled through the resonant characteristics (i.e. equal resonance bandwidths) of the ring cavity in combination with the excitation scheme described above. Setting the pump power to 1.5 mW, a quadruple detection rate of 0.17 Hz was measured, leading to a calculated generation rate of 135 kHz – by taking into account the system and detection losses of 14.75 dB. We then performed four-photon quantum interference measurements (Fig. 3F). Note that four-photon interference is in general not present for two completely independent two-photon qubit states. The interference is expected to be proportional to $3 + cos(4\alpha - 4\varphi) + 4cos(2\alpha - 2\varphi)$, where α is the phase of all four entangled photons and φ the pump interferometer phase (*21*). Our data clearly follows the expected relation, having a visibility of 89% without compensation for background noise or losses. Furthermore, we repeated the four-photon measurement by selecting different combinations of four modes, always finding four-photon entanglement, see Fig. S3 and (*21*) for more details.

Finally, to fully characterize the entangled states we performed quantum state tomography (*30*). This method measures the state density matrix, from which it is possible to extract important characteristics such as the fidelity, which describes how close the measured state is to the ideal entangled state (*21*). We first measure the two-photon qubits generated on comb lines symmetric with respect to the pump wavelength (Fig. 4A,B) and find a fidelity of 96%, confirming that the generated quantum states are of high quality and very close to the ideal entangled state. For the four-photon entangled state (Fig. 4C,D), we obtained a fidelity of 64% without compensation for background noise or interferometer imperfections, which is comparable to the fidelity measured for non-integrated four-photon states used for practical applications (*3*).

A key characteristic of our quantum frequency comb is the intrinsic and simultaneous operation at many modes, the high-purity photon pairs as well as high quality bi- and multi-photon entanglement shared among these modes, and an intrinsic compatibility with fiber technology. Due to these features, it can find versatile and immediate applications such as quantum communications and quantum computation. For example, the source can be implemented into both single-photon as well as entanglement-based quantum communication protocols. The broadband nature of the quantum comb is particularly attractive for multi-channel applications, where the amount of transmitted data can be increased through the use of multiple, equally well-performing channels. We repeated the two-photon tomography measurement after adding 40 km of fiber, measuring a fidelity of 87% (Fig. S4, and (*21*) for more details), demonstrating that the entanglement is still preserved after long fiber propagation.

Furthermore, two-photon time-bin entangled qubits have successfully been used for linear universal quantum computation (*4*, *25*), and the parallel generation and processing of multiple qubits can directly enhance such protocols, where the information capacity scales with the number of comb lines used, as theoretically predicted (*25*).

Even though the demonstrated multi-photon entangled states are separable, as they are generated as a product of bi-photon Bell states, it is conceivable that through the use of multiple excitation fields (*6*) or controlled phase gates (*31*), non-separable multi-photon cluster states could be constructed. Indeed, four-photon cluster states have been used to demonstrate measurement-based quantum computation (*3*).

Further device integration of the frequency comb will lead to more compact and stable systems with higher performance in terms of detection rates. All components used in our setup, such as the laser, filters, interferometers and detectors (here connected via optical fibers), could be integrated on a single chip (*13*) to reduce size and losses (currently at 14.75 dB). A very realistic decrease in losses by 5 dB would already increase the four-photon detection rate by a factor of one hundred, while an achievable loss reduction of 10 dB would increase it to the very useful kHz range.

Our results indicate that integrated quantum frequency comb sources based on third-order nonlinearities can open up new venues for the generation and control of complex quantum states, thus providing a scalable and practical platform for optical quantum information processing.

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Supplementary Material
Materials and methods
Supplementary text
Figures S1 to S4
References (32-37)

Fig. 1: Measured single photon spectrum of the integrated quantum frequency comb. Single photon spectrum emitted by the microring resonator, measured using a grating-based spectrum analyzer and a high resolution digital tunable filter in the C-Band (bottom left inset). For clarity the S, C and L telecommunication bands are also indicated in the figure. The blue curve shows the spectral asymmetry, which can be explained by Raman scattering. The channels used in the entanglement measurements are shown in the left bottom inset, where the measured raw (and background corrected, in parentheses) entanglement visibilities for the individual channel pairs are shown in the top left inset.

Fig. 2: Quantum frequency comb. A pulsed laser is passed through an unbalanced fiber Michelson interferometer, generating double pulses with a phase difference $\boldsymbol{\varphi}$. The pulses are fed into the microring resonator, exciting one microring resonance, generating time-bin entangled photon pairs on a frequency comb through SFWM. For analysis purposes (entanglement verification – see Fig. 3A-D or quantum state tomography – see Fig. 4A-B), each photon of the spectrally filtered photon pair is individually passed through an interferometer with the temporal imbalance equal to the time slot separation, and then detected with a single photon detector. For the four-photon measurements (see Fig. 3E and Fig. 4C-D), four frequency modes symmetric to the excitation field are collected and passed through the interferometers.

Fig. 3: Entanglement and phase control via SFWM. To demonstrate the difference between the phase characteristics of SPDC and SFWM, five different quantum interference measurements are performed. Three interferometer phases are adjusted: $\boldsymbol{\varphi}, \boldsymbol{\alpha}, \boldsymbol{\beta}$ being the phases of the pump, signal and idler interferometers, respectively. A) $\boldsymbol{\varphi = 0}, \boldsymbol{\alpha = \beta} \in [\mathbf{0}, \mathbf{2\pi}]$, B) $\boldsymbol{\varphi} \in [\mathbf{0}, \mathbf{2\pi}]$, $\boldsymbol{\alpha = \beta = 0}$, C) $\boldsymbol{\alpha} \in [\mathbf{0}, \mathbf{2\pi}], \boldsymbol{\varphi = \beta = 0}$, D) $\boldsymbol{\alpha = \beta = \varphi} \in [\mathbf{0}, \mathbf{2\pi}]$, E) $\boldsymbol{\alpha = \beta = -\varphi} \in [\mathbf{0}, \mathbf{2\pi}]$. The error bars represent the standard deviation of 7 measurements. F) Four-photon entanglement measurement with all photon phases tuned simultaneously, showing clear four-photon quantum interference with a visibility of 89%. The solid line indicates the expected function, while the dashed line shows the cosine interference in the two-photon case.

Fig. 4: Quantum state tomography measurement. Ideal (**A**) and measured **(B)** density matrix of two-photon time-bin entangled qubits, revealing a state fidelity of 96%. Ideal (**C)** and measured **(D)** density matrix of four-photon time-bin entangled qubits, revealing a state fidelity of 64%.