Frequency-bin entanglement from domain-engineered down-conversion

Cite as: APL Photonics 7, 066102 (2022); https://doi.org/10.1063/5.0089313
Submitted: 24 February 2022 • Accepted: 13 May 2022 • Published Online: 01 June 2022

Christopher L. Morrison, Francesco Graffitti, Peter Barrow, et al.

COLLECTIONS

This paper was selected as an Editor’s Pick

ARTICLES YOU MAY BE INTERESTED IN

Optical parametric wideband frequency modulation
APL Photonics 7, 066106 (2022); https://doi.org/10.1063/5.0092969

Dual-mode microresonators as straightforward access to octave-spanning dissipative Kerr solitons
APL Photonics 7, 066103 (2022); https://doi.org/10.1063/5.0089036

Hybrid integrated external cavity laser with a 172-nm tuning range
APL Photonics 7, 066101 (2022); https://doi.org/10.1063/5.0088119
Frequency-bin entanglement from domain-engineered down-conversion

Cite as: APL Photon. 7, 066102 (2022); doi: 10.1063/5.0089313
Submitted: 24 February 2022 • Accepted: 13 May 2022 • Published Online: 1 June 2022

Christopher L. Morrison, Francesco Graffitti, Peter Barrow, Alexander Pickston, Joseph Ho, and Alessandro Fedrizzi

AFFILIATIONS
Institute of Photonics and Quantum Sciences, School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom

ABSTRACT
Frequency encoding is quickly becoming an attractive prospect for quantum information protocols owing to larger Hilbert spaces and increased resilience to noise compared to other photonic degrees of freedom. To fully make use of frequency encoding as a practical paradigm for quantum information processing, an efficient and simple source of frequency entanglement is required. Here, we present a single-pass source of discrete frequency-bin entanglement that does not use filtering or a resonant cavity. We use a domain-engineered nonlinear crystal to generate an eight-mode frequency-bin entangled source at telecommunication wavelengths. Our approach leverages the high heralding efficiency and simplicity associated with bulk crystal sources.

© 2022 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/). https://doi.org/10.1063/5.0089313

I. INTRODUCTION
Quantum photonics is increasingly exploiting the time-frequency degree of freedom (DoF) due to its compatibility with telecom infrastructure, the potential for greater information capacity per photon, and improved resilience to noise.1,2 Time-frequency encodings can be broadly divided into three categories: time-bin encoded, frequency-bin encoded, and intensity-nonorthogonal "pulse-mode" encoded states (see Fig. 1).

Time-bin states are encoded in discrete arrival times at the detection apparatus with a time separation exceeding the duration of the time-bins. Time-bin experiments date back several decades, and the encoding is easily manipulated with unbalanced interferometers and phase shifters and detected using standard single-photon detectors.3,4 The complement of time-bin encoding is frequency-bin encoding, with photons localized in non-overlapping spectral regions.3,4 Frequency-bin states can be manipulated and detected using electro-optic modulators5 and Fourier transform pulse shapers which can, in principle, be lossless.3 This has allowed for full tomography of frequency-bin entangled photon pairs5,6 and the generation of on-chip cluster states.7,8 Pulse-mode encoded states are the most recent addition to the experimental time-frequency toolbox, having initially been studied in the context of spectral entanglement in pulsed parametric down-conversion (PDC).9

Pulse-modes are overlapping in both spectral and temporal amplitude, and their orthogonality is maintained by considering the temporal or spectral phase between different states. Due to their intensity nonorthogonality, pulse-modes offer a way to extend time-frequency encoding beyond purely time or frequency-bin states (see Fig. 1). The introduction of the quantum pulse gate13 has made it possible to arbitrarily manipulate and measure pulse-modes. In this work, we focus on the generation of frequency-bin encoded states.

Discrete frequency-bin entangled states have been generated by filtering broadband correlated biphotons,14,15 which are attractive due to their compatibility with commercial wavelength-division multiplexing (WDM) telecom components. Frequency-bin entanglement can also be generated on-chip by using microresonators that produce photon pairs across multiple cavity resonances—an intrinsically stable approach that can be scaled to a large number of sources on one chip. The main drawback of both these approaches is optical loss due to filtering and/or resonant losses inside the microresonators, which drastically limits the achievable heralding efficiency. Techniques from ultrafast pulse shaping can also be used to generate time-frequency encoded states16 but arguably a simpler technique is the use of PDC with domain-engineered nonlinear...
 crystals, which allows for the tailoring of almost arbitrary biphoton phasematching functions (PMF). Domain-engineered crystals have been used to generate Gaussian biphoton states for pure heralded single photons and maximally entangled pulse-modes in two dimensions. Dual-poled crystals have also been used to generate frequency-bin entanglement over two modes.

In this work, we demonstrate an eight-mode frequency-bin entangled source centered at telecom wavelengths. We use a single pass of a pump beam to generate the frequency-bin entanglement without the need for filtering. This process is, in principle, lossless and can allow for high heralding efficiencies. This is in contrast to typical frequency-bin entangled sources, which are inherently lossy due to filtering or the use of an optical cavity, typically around 3–7 dB total loss. Typical frequency-bin sources require continuous-wave (CW) pumping; our approach uses pulsed excitation, which can be scaled up for use in multiphoton protocols, as demonstrated by heralded two-photon interference measurements. We verify eight-mode frequency entanglement through a combination of joint spectral intensity (JSI) and two-photon interference measurements. Using the crystal inside a Sagnac interferometer, we also demonstrate polarization-frequency hyperentanglement with frequency-resolved polarization tomography.

II. RESULTS AND DISCUSSION

Our domain-engineering algorithm tracks a target PDC amplitude along the crystal and chooses the domain width and orientation on a domain-by-domain basis to maximize the overlap to the desired target PMF. The target PMF, in this work, was a comb of eight separate Gaussian peaks (see the supplementary material). The crystal was manufactured by Raicol Ltd. With the symmetric group-velocity matching condition in type-II PDC in potassium...
titanyl phosphate (KTP), the eight Gaussian peaks in the PMF function result in a spectrally entangled state. Using the Schmidt decomposition, the biphoton state can be written as

$$\ket{\psi} = \sum_i \sqrt{\lambda_i} A_i^\dagger B_i^\dagger \ket{00},$$

where $A_i^\dagger, B_i^\dagger$ are broadband mode operators and $\lambda_i$ are the Schmidt coefficients.\textsuperscript{12} For a maximally entangled eight-mode state, $\lambda_i = 1/8$. The Schmidt number $K = 1/\sum \lambda_i^2$ defines the effective number of modes in the PDC process, for an $n$-mode maximally entangled state $K = n$.

In this experiment, the eight-bin KTP crystal (8b-KTP) is pumped by a Ti-sapphire laser with a base repetition rate of 80 MHz, a pulse duration of 1.3 ps, and a central wavelength of 777.85 nm, which leads to degenerate photons centred at 1555.7 nm. The width of the frequency bins is chosen to maximize the overlap with an eight-mode maximally entangled state for the available pump pulse duration. The pump is focused into the crystal with a 40 cm focal length lens for a beam waist of ~77 μm. The pump light is removed with a silicon filter and a long pass filter at 1400 nm before the photons are collected into single-mode fibers. The photons are detected with superconducting nanowire single-photon detectors (SNSPDs) with a nominal quantum efficiency of 80% and a jitter of 50 ps. Time-tags are recorded using a single-photon spectrometer with a silicon filter and a long pass filter at 1400 nm before the photons are detected with a silicon filter and a long pass filter at 1400 nm before the photons are detected.

We measure the joint spectral intensity of the biphoton state using a dispersive fiber spectrometer [see Fig. 2(b)]. The fiber dispersion maps the frequency of the photons to the arrival time at the detectors. This allows for a simple reconstruction of the joint spectral intensity using coincidence counts. Reconstructing the joint spectral amplitude with full spectral phase information is possible\textsuperscript{24,27,28} but typically requires well characterized reference beams or electro-optic effects. We probe possible spectral phase correlations, which would increase the number of modes present in the down-conversion process, using heralded two-photon interference measurements, and show that the visibility is consistent with an eight-mode maximally entangled state.

We numerically calculate the Schmidt decomposition of the square root of the joint spectral intensity from $4.3 \times 10^7$ detected events. We calculate an overlap of 96.01(1)% to an eight-mode maximally entangled state by comparing Schmidt numbers to the ideal value of $\lambda_i = 1/8$ for the first eight Schmidt modes and zero otherwise. We also estimate a Schmidt number of $K = 7.018(3)$, and both errors are quoted at three sigmas estimated from 1000 rounds of Monte Carlo simulation assuming Poissonian counting statistics. The theoretical values based on the designed phasematching function are 98.5% and 8.07, respectively. The overlap between the theoretical design and measured values is 97.47(1)%. We attribute the difference in Schmidt number and fidelity to the unequal peak heights and non-zero extinction between the peaks, most clearly seen in the marginals of Fig. 3. The variation in peak height is most likely due to different detection efficiencies for each bin. As the photons propagate along the 20 km fibers, each bin experiences a different birefringence due to the refractive index variation with wavelength, resulting in a different polarization state for each bin at the SNSPDs, which are polarization sensitive. The average separation between peaks is measured to be 498 GHz, which matches well with the designed separation of 500 GHz. The JSI is calibrated against marginal spectra measurements with a commercial single-photon spectrometer.

In order to assess phase correlations in the joint spectrum, we also carry out heralded two-photon interference. For an $n$-mode maximally entangled PDC process, the heralded interference visibility is given by $1/n$; therefore, we expect a visibility of 12.5% for an eight-mode entangled state. We measure an interference...
FIG. 4. Theoretical (a) and experimentally measured (b) joint spectral intensity. The JSI is reconstructed through TOFS on a 500 × 500 grid with each pixel corresponding to an arrival window of 25 ps, which corresponds to a spectral resolution of around 0.06 nm. The spectral range of the spectrometer is set by the temporal separation between pulses from the pump laser corresponding to 12.5 ns, which is wide enough to contain all eight frequency-bins. The number of frequency-bins could be extended at the cost of reducing the repetition rate of the pump laser. We include the marginal distributions on the left and top of the main JSI plot, with counts normalized to the maximum peak height.

visibility of 11.2(1.4)% (see the supplementary material), which agrees with the expected visibility within experimental error. The discrepancy between the measured and theoretical visibility is attributed to small variations in the polarization state across the full bandwidth of the photons due to chromatic dispersion in the fiber polarization controllers. A mixed state with no spectral phase coherence would produce the same visibility in the heralded two-photon interference measurements but would not be consistent with the measured biphoton interference. Using the crystal in a Sagnac interferometer, we generate a hyperentangled state in polarization and frequency mode of the form

$$|\psi\rangle = \frac{1}{\sqrt{N_i}} \sum_{i=-4}^{4} (|H\rangle|V\rangle - e^{i\phi_i}|V\rangle|H\rangle) \otimes |i\rangle - |i\rangle,$$

with $N_i$ related to the probability of emitting a pair of photons into the $i$-th frequency bin. Domain-engineered crystals have previously been used inside a Sagnac interferometer to demonstrate hyperentanglement between polarization and pulse-mode.\(^{29}\) Due to the wavelength dependent retardance of the polarization optics within the Sagnac source, each frequency-bin has a different phase $\phi_i$. We show that the polarization entanglement persists across all frequency-bins by using TOFS after polarization tomography. The TOFS measurement mimics the action of a WDM by temporally demultiplexing the frequency components into different arrival times. We perform quantum state tomography using symmetric informationally complete (SIC) projections on the two-qubit polarization state. This allows us to use fewer measurements when projecting with a single detector outcome; see the supplementary material for more details.

The fidelity to the singlet state is calculated by applying a correction for the phases $\phi_i$ in post-processing. In a wavelength-division multiplexed scenario, this phase can be corrected by appropriate waveplate settings. The average purity and fidelity are 88.7(3)% and 92.6(1)%, respectively, with a maximum (minimum) fidelity of 97.3% (88.7%); see the supplementary material for more details. Errors are calculated by 1000 rounds of Monte Carlo simulation assuming Poissonian counting statistics. The drop in purities when compared to other telecom wavelength Sagnac sources\(^{30}\) is attributed to the wavelength dependence of the polarization optics inside the Sagnac interferometer and can be improved by using achromatic optics. The waveplates in the tomography setup also impart different unitaries across the full bandwidth of the downconverted photons, which reduces the purity of the reconstructed state.

This can be mitigated by reconstructing the polarization state of each bin individually using WDM filters\(^{31}\) or again using achromatic optics.

III. CONCLUSION

The efficient, pulsed source of frequency-bin entanglement presented in this work can be extended and improved in future work in multiple ways. Improving crystal fabrication to match lengths available with other materials such as lithium niobate would allow for denser packing of spectral features using domain engineering. With a longer crystal, the frequency-bin spacing can be made to more closely match the International Telecommunication Union (ITU) standard 100 GHz grid, which will allow for efficient integration with future WDM quantum networks.\(^{32,33}\) As the frequency-bin entanglement is generated with pulsed excitation, it would be possible to efficiently carry out Bell state measurements and, therefore, entanglement swapping in a WDM network. This would provide a way to connect users across initially unconnected WDM networks using a central node to carry out Bell state measurements.
The domain engineering technique used here could be combined with other methods of shaping the joint spectrum, such as pump shaping, to produce time-frequency grid states or using time-frequency synthesis techniques to double the number of spectral features produced from domain engineering. An open question for future work is if domain engineering can be used to generate biphotons in hybrid encodings (see Fig. 1) with a combination of pulse-modes, time-bins, or frequency-bins, which could be used to directly generate cluster states in the time-frequency degree of freedom.

SUPPLEMENTARY MATERIAL

The supplementary material contains information on the design of the frequency-bin crystal, the nonlinearity profile, and the phase-matching function alongside calculations for the interference patterns from the joint spectrum. It also contains the heralded two-photon interference data and the polarization and frequency-bin hyperentanglement data.

ACKNOWLEDGMENTS

The authors thank B. D. Gerardot and M. Malik for the loan of equipment. This work was supported by the UK Engineering and Physical Sciences Research Council (Grant No. EP/T001011/1). F.G. acknowledges studentship funding from EPSRC under Grant No. EP/L015110/1.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

1. Y. Ding, D. Bacco, K. Dalgaard, X. Cai, X. Zhou, K. Rottwitt, and L. K. Oxenløwe, “High-dimensional quantum key distribution based on multicore fiber using silicon photonic integrated circuits,” npj Quantum Inf. 3, 25 (2017).
2. S. Ecker, F. Bouchard, L. Bulla, F. Brandt, O. Kohout, F. Steinlechner, R. Pickler, M. Malik, Y. Guryanova, R. Ursin, and M. Huber, “Overcoming noise in entanglement distribution,” Phys. Rev. X 9, 041042 (2019).
3. J. Brendel, N. Gisin, W. Tittel, and H. Zbinden, “Pulsed energy-time entangled twin-photon source for quantum communication,” Phys. Rev. Lett. 82, 2594–2597 (1999).
4. I. Marcikic, H. de Riedmatten, W. Tittel, V. Scarani, H. Zbinden, and N. Gisin, “Time-bin entangled qubits for quantum communication created by femtosecond pulses,” Phys. Rev. A 66, 062308 (2002).
5. S. Ramelow, L. Ratschbacher, A. Fedrizzi, N. K. Langford, and A. Zeilinger, “Discrete tunable color entanglement,” Phys. Rev. Lett. 103, 253601 (2009).
6. L. Olsislager, J. Cussey, A. T. Nguyen, P. Emplit, S. Massar, J.-M. Merolla, and K. P. Huy, “Frequency-bin entangled photons,” Phys. Rev. A 82, 013804 (2010).
7. L. Olsislager, E. Woodhead, K. Phan Huy, J.-M. Merolla, P. Emplit, and S. Massar, “Creating and manipulating entangled optical qubits in the frequency domain,” Phys. Rev. A 89, 052323 (2014).
8. H.-H. Lu, E. M. Simmerman, P. Lougovski, A. M. Weiner, and J. M. Lukens, “Fully arbitrary control of frequency-bin qubits,” Phys. Rev. Lett. 125, 12503 (2020).
9. M. Kues, C. Reimer, P. Rottocci, L. Cortés, S. Sciara, B. Wetzel, Y. Zhang, A. Cino, S. T. Chu, B. E. Little, D. J. Moss, L. Caspani, J. Azaña, and R. Morandotti, “On-chip generation of high-dimensional entangled quantum states and their coherent control,” Nature 546, 622–626 (2017).
10. H.-H. Lu, K. V. Myllyswn, R. S. Bennink, S. Seshadri, M. S. Al-Ashkhy, J. Liu, T. J. Kippenberg, D. E. Leaird, A. M. Weiner, and J. M. Lukens, “Full quantum state tomography of high-dimensional on-chip biphoton frequency combs with randomized measurements,” arXiv:2108.04124 [quant-ph] (2021).
11. C. Reimer, S. Sciara, P. Rottocci, M. Islam, L. Romero Cortés, Y. Zhang, B. Fischer, S. Loranger, R. Kashyap, A. Cino, S. T. Chu, B. E. Little, D. J. Moss, L. Caspani, W. J. Munro, J. Azaña, M. Kues, and R. Morandotti, “High-dimensional one-way quantum processing implemented on d-level cluster states,” Nat. Phys. 15, 148–153 (2019).
12. C. K. Law, I. A. Walmsley, and J. H. Eberly, “Continuous frequency entanglement: Effective finite Hilbert space and entropy control,” Phys. Rev. Lett. 84, 5304–5307 (2000).
13. A. Eckstein, B. Brecht, and C. Silberhorn, “A quantum pulse gate based on spectrally engineered sum frequency generation,” Opt. Express 19, 13779–13778 (2011).
14. K.-C. Chang, X. Cheng, M. C. Sarhan, A. K. Vinod, Y. S. Lee, T. Zhong, Y.-X. Gong, Z. Xie, J. H. Shapiro, F. N. C. Wong, and C. W. Wong, “648 Hilbert-space dimensionality in a biphoton frequency comb: Entanglement of formation and Schmidt mode decomposition,” npj Quantum Inf. 7, 48 (2021).
15. N. B. Lingaraju, H.-H. Lu, S. Seshadri, P. Imany, D. E. Leaird, J. M. Lukens, and A. M. Weiner, “Quantum frequency combs and Huo-On-Mandel interference: The role of spectral phase coherence,” Opt. Express 27, 38683–38697 (2019).
16. V. Ansari, J. M. Donohue, B. Brecht, and C. Silberhorn, “Tailoring nonlinear processes for quantum optics with pulsed temporal-mode encodings,” Optica 5, 534–550 (2018).
17. F. Graffitti, D. Kundys, D. T. Reid, A. M. Braiczyk, and A. Fedrizzi, “Pure down-conversion photons through sub-coherence-length domain engineering,” Quantum Sci. Technol. 2, 035001 (2017).
18. F. Graffitti, P. Barrow, M. Proietti, D. Kundys, and A. Fedrizzi, “Independent high-purity photons created in domain-engineered crystals,” Optica 5, 514–517 (2018).
19. A. Pickston, F. Graffitti, P. Barrow, C. L. Morrison, J. Ho, A. M. Braiczyk, and A. Fedrizzi, “Optimised domain-engineered crystals for pure telecom photon sources,” Opt. Express 29, 6991–7002 (2021).
20. F. Graffitti, P. Barrow, A. Pickston, A. M. Braiczyk, and A. Fedrizzi, “Direct generation of tailored pulse-mode entanglement,” Phys. Rev. Lett. 124, 053603 (2020).
21. F. Kaneda, H. Suzuki, R. Shimizu, and K. Edamatsu, “Direct generation of frequency-bin entangled photons via two-period quasi-phase-matched parametric downconversion,” Opt. Express 27, 1416–1424 (2019).
22. Z. Xie, T. Zhong, S. Shrestha, X. Xu, J. Liang, Y.-X. Gong, J. C. Bienfang, A. Restelli, J. H. Shapiro, F. N. C. Wong, and C. Wei Wong, “Harnessing high-dimensional hyperentanglement through a biphoton frequency comb,” Nat. Photonics 9, 536–542 (2015).
23. C. Reimer, M. Kues, P. Rottocci, B. Wetzel, F. Grazioso, B. E. Little, S. T. Chu, T. Johnston, Y. Bromberg, L. Caspani, D. J. Moss, and R. Morandotti, “Generation of multiphoton entangled quantum states by means of integrated photon sources,” Science 351, 1176–1180 (2016).
24. A. O. C. Davis, V. Thiel, and B. J. Smith, “Measuring the quantum state of a photon pair entangled in frequency and time,” Optica 7, 1317–1322 (2020).
25. A. Fedrizzi, T. Herbst, M. Aspelmeyer, M. Barbieri, T. Jennewein, and A. Zeilinger, “Anti-symmetrization reveals hidden entanglement,” New J. Phys. 11, 103052 (2009).
26 M. A. Broome, M. P. Almeida, A. Fedrizzi, and A. G. White, "Reducing multi-photon rates in pulsed down-conversion by temporal multiplexing," Opt. Express 19, 22698–22708 (2011).

27 I. Jizan, B. Bell, L. G. Helt, A. C. Bedoya, C. Xiong, and B. J. Eggleton, "Phase-sensitive tomography of the joint spectral amplitude of photon pair sources," Opt. Lett. 41, 4803–4806 (2016).

28 G. Triginer, M. D. Vidrighin, N. Quesada, A. Eckstein, M. Moore, W. S. Kolthammer, J. Sipe, and I. A. Walmsley, "Understanding high-gain twin-beam sources using cascaded stimulated emission," Phys. Rev. X 10, 031063 (2020).

29 F. Graffitti, V. D’Ambrosio, M. Proietti, J. Ho, B. Piccirillo, C. de Lisio, L. Marrucci, and A. Fedrizzi, "Hyperentanglement in structured quantum light," Phys. Rev. Res. 2, 043350 (2020).

30 R.-B. Jin, R. Shimizu, K. Waku, M. Fujiwara, T. Yamashita, S. Miki, H. Terai, Z. Wang, and M. Sasaki, "Pulsed Sagnac polarization-entangled photon source with a PPKTP crystal at telecom wavelength," Opt. Express 22, 11498–11507 (2014).

31 S. P. Neumann, M. Selimovic, M. Bohmann, and R. Ursin, "Experimental entanglement generation for quantum key distribution beyond 1 Gbit/s," arXiv:2107.07736 [quant-ph] (2021).

32 S. Wengerowsky, S. K. Joshi, F. Steinlechner, H. Hübel, and R. Ursin, "An entanglement-based wavelength-multiplexed quantum communication network," Nature 564, 225–228 (2018).

33 S. K. Joshi, D. Aktas, S. Wengerowsky, M. Lončarić, S. P. Neumann, B. Liu, T. Scheidl, G. C. Lorenzo, Ž. Samec, L. Kling, A. Qiu, M. Razavi, M. Stipčević, J. G. Rarity, and R. Ursin, "A trusted-node-free eight-user metropolitan quantum communication network," Sci. Adv. 6, eaba0959 (2020).

34 N. Fabre, G. Maltese, F. Appas, S. Felicetti, A. Ketterer, A. Keller, T. Coudreau, F. Bahoux, M. I. Amanti, S. Ducci, and P. Milman, "Generation of a time-frequency grid state with integrated biphoton frequency combs," Phys. Rev. A 102, 012607 (2020).

35 R.-B. Jin, K. Tazawa, N. Asamura, M. Yabuno, S. Miki, F. China, H. Terai, K. Minoshima, and R. Shimizu, "Quantum optical synthesis in 2D time–frequency space," APL Photonics 6, 086104 (2021).