Full multipartite entanglement of frequency comb Gaussian states

S. Gerke,1 J. Sperling,1 W. Vogel,1 Y. Cai,2 J. Roslund,2 N. Treps,2 and C. Fabre2

1Arbeitsgruppe Theoretische Quantenoptik, Institut f¨ur Physik, Universit¨at Rostock, D-18051 Rostock, Germany
2Laboratoire Kastler Brossel, Sorbonne Universit´es - UPMC, ´Ecole Normale Sup´erieure, Coll`ee de France, CNRS; 4 place Jussieu, 75252 Paris, France

(Dated: September 22, 2014)

An analysis is conducted of the multipartite entanglement for Gaussian states generated by the parametric downconversion of a femtosecond frequency comb. Using a recently introduced method for constructing optimal entanglement criteria, a family of tests is formulated for mode decompositions that extend beyond the traditional bipartition analyses. A numerical optimization over this family is performed to achieve maximal significance of entanglement verification. For experimentally prepared 4, 6, and 10-mode states, full entanglement is certified for all of the 14, 202, and 115974 possible nontrivial partitions, respectively.

I. INTRODUCTION

One of the most fundamental concepts in quantum physics is entanglement, which may be described as a quantum correlation between compound systems [1]. This property of entanglement plays a central role in a host of quantum technologies, including metrology, imaging, communication, and quantum information processing [2,4]. Protocols in each of these domains rely upon the distribution of nonclassical correlations among a multitude of subsystems within a state [5,7]. As such, reliable, readily-implementable, and versatile means of characterizing entanglement are essential for assessing the utility of certain states as well as understanding the fundamental physics underlying quantum interactions.

One method for identifying entanglement is formulated in terms of positive, but not completely positive maps. The most prominent example of such a map is the partial transposition (PT) criterion [8]. For bipartite Gaussian states, which are completely characterized by the covariance matrix, it has been shown that the PT criterion is necessary and sufficient to identify entanglement [9,10]. In the multipartite case, however, the PT criteria can only diagnose entanglement among state bipartitions. Moreover, bound entangled Gaussian states are known to exist whose entanglement cannot be detected with the PT criterion. Such states have been formulated in theory and also realized in experiments [11,12]. Additionally, a number of moment-based entanglement probes have been successfully deployed to characterize the entanglement of a state, e.g., [13,22].

These criteria have been enormously successful at experimentally diagnosing entanglement among various beams [23,24] or among different parties of a multimode beam [25,26]. Alternatively, several studies have acquired the covariance matrix for a multidimensional state, which enables implementation of the PT criterion as a means for examining the nonclassical correlations among multiple beams [27,28]. In each of these situations, however, the employed methods restrict multipartite dynamics to the set of all possible bipartite state divisions.

Another well-established method for identifying entanglement is formulated in terms of entanglement witnesses [29,30]. In particular, the separability eigenvalue equations have recently been introduced as a method for constructing optimal witnesses [31,32]. The solutions of these coupled equations yield powerful entanglement assessments not only for bipartite divisions but also for high-order multipartite divisions of discrete and continuous variable quantum systems.

This contribution formulates entanglement conditions for multimode Gaussian states and subsequently demonstrates their application on an experimentally realized quantum ultrafast frequency comb. This quantum state, which is generated by the parametric downconversion of a classical frequency comb, was recently shown to exhibit entanglement among all of its underlying frequencies [33]. The covariance matrix for this high-dimensional quantum object has been measured, which renders it a unique testbed for exploring novel multipartite entanglement metrics. Importantly, the criteria developed from the separability eigenvalue equations are able to examine nonclassical aspects of the frequency comb not feasible with strictly bipartite methods. Within this class of criteria, the significance of the verified entanglement is optimized with a genetic algorithm, which allows us to fully verify the entanglement present in highly complex multiparty quantum systems. For the 10-mode system considered here, entanglement is certified for each of the 115974 possible nontrivial state partitions.

II. GAUSSIAN STATES AND MODE DECOMPOSITIONS

Gaussian states are described by a Gaussian characteristic function on a multimode phase space (for an intro-
duction see, e.g., [34]). The amplitude and phase quadratures of individual modes are denoted \( \hat{x}_k \) and \( \hat{p}_k \), respectively, and a vector of quadratures is defined as
\[
\hat{\xi} = (\hat{x}_1, \ldots, \hat{x}_N, \hat{p}_1, \ldots, \hat{p}_N)^T.
\]
(1)
The covariance matrix \( C \) is then specified by its entries
\[
C^{ij} = \frac{1}{2} (\langle \xi_i \xi_j \rangle + \langle \xi_j \xi_i \rangle) - \langle \xi_i \rangle \langle \xi_j \rangle.
\]
(2)
First-order moments are irrelevant for entanglement since local unitary displacement operations may be applied to the state to yield \( \langle \xi \rangle = 0 \). Thus, without loss of generality, we can assume that all of the information for a Gaussian state is contained in its second-order moments.

A quantum state consisting of \( N \) modes is then decomposed into a set of \( K \) partitions: \( I_1, \ldots, I_K \). The state is considered as entangled with respect to this mode partitioning if one is not able to write it as a classical mixture of product states \( |a_1, \ldots, a_K \rangle \), with \( |a_j \rangle \in H_j \) being a vector of the subsystem \( I_j \). For example, the set corresponding to \( K = 2 \) encapsulates all state bipartitions, which corresponds to those partitions addressed by the PT criterion. However, even if entanglement does not exist among certain bipartitions, it may be present in higher-order partitions, i.e., \( K > 2 \). Considering that the total number of state partitions is given by the Bell number and increases rapidly as a function of \( N \) [35], the PT criterion addresses only a very small subset of the rich variety of possible partitions.

### III. OPTIMAL ENTANGLEMENT TESTS

The multipartite entanglement of a quantum state \( \hat{\rho} \) may be probed with the use of a general Hermitian operator \( \hat{L} \) [31]. In particular, the state under question is entangled with respect to a given \( K \)-partition if and only if it may be shown that
\[
\text{tr}(\hat{L} \hat{\rho}) < \min_{I_1: \ldots: I_K} g_{I_1: \ldots: I_K}^\text{min},
\]
(3)
where \( g_{I_1: \ldots: I_K}^\text{min} \) is the minimum expectation value of \( \hat{L} \) among all of the states of the \( K \)-party subsystem under the assumption that they are separable. It was established in [31] that this minimization problem can be solved with a set of coupled eigenvalue equations, denoted as separability eigenvalue equations. The resulting minimal separability eigenvalue is identical to \( g_{I_1: \ldots: I_K}^\text{min} \).

The most general form of the operator \( \hat{L} \) for continuous variable Gaussian states is given as
\[
\hat{L} = \sum_{i,j} \left( M_{xx}^{ij} \hat{x}_i \hat{x}_j + M_{xp}^{ij} \hat{x}_i \hat{p}_j + M_{px}^{ij} \hat{p}_i \hat{x}_j + M_{pp}^{ij} \hat{p}_i \hat{p}_j \right)
\]
in which the coefficients of \( M \) are freely adjustable. Accordingly, attention may be restricted to the state’s covariance matrix. Correlations between the amplitude and phase quadratures are negligible for the presently studied states, which allows the test operator \( \hat{L} \) to be cast as
\[
\hat{L} = \text{Tr}(M \hat{\xi} \hat{\xi}^T), \quad \text{with } M = \begin{pmatrix} M_{xx} & 0 \\ 0 & M_{pp} \end{pmatrix} = M^T > 0,
\]
(4)
where \( M_{xx} \) and \( M_{pp} \) are coefficient matrices of the same dimensionality as the corresponding state covariance matrix, and the indices \( xx \) and \( pp \) refer to amplitude-amplitude and phase-phase correlations, respectively. The expectation value of this test operator readily follows and is written as
\[
\langle \hat{L} \rangle = \text{tr}(\hat{L} \hat{\rho}) = \text{Tr}(MC).
\]
(5)
Likewise, the minimal separability eigenvalue \( g_{I_1: \ldots: I_K}^\text{min} \) for operators of this form has been derived in Ref. [31] and reads as
\[
g_{I_1: \ldots: I_K}^\text{min} = \sum_{k=1}^{K} \text{Tr}( \left[ M_{pp}^{1/2} I_{xx}, I_{xx}, I_{xx}, M_{pp}^{1/2} \right] )^{1/2},
\]
(6)
where \( M_{Ik} \) are the submatrices of \( M \) that contain only the rows and columns of the modes within \( I_k \).

A partition’s entanglement is characterized in terms of its statistical significance \( \Sigma \), which compares the difference between the expectation value \( \langle \hat{L} \rangle \) and its separable bound \( g_{I_1: \ldots: I_K}^\text{min} \) to the experimental standard deviation \( \sigma(\hat{L}) \):
\[
\Sigma = \frac{\langle \hat{L} \rangle - g_{I_1: \ldots: I_K}^\text{min}}{\sigma(\hat{L})},
\]
(7)
which is the considered entanglement metric. The experimental error \( \sigma(\hat{L}) \) is determined through error propagation of \( \langle \hat{L} \rangle \) and yields
\[
\sigma(\hat{L}) = \sqrt{\sum_{i,j=1}^{N} (\sigma(C_{xx}^{ij}))^2 + (\sigma(C_{pp}^{ij}))^2},
\]
(8)
where \( \sigma(C_{xx}^{ij}) \) and \( \sigma(C_{pp}^{ij}) \) are the measured errors corresponding to the covariance elements \( C_{xx}^{ij} \) and \( C_{pp}^{ij} \), respectively. A partition is considered entangled if \( \Sigma < 0 \), and the statistical significance of its non-separability is assessed with \( |\Sigma| \). The coefficient matrix \( M \) may be freely tuned in order to maximize the significance of each partition, \( \Sigma \rightarrow \Sigma_{\text{min}} < 0 \). This optimization is achieved with a genetic algorithm.

### IV. EXPERIMENTAL REALIZATION

Femtosecond frequency combs contain upwards of \( \sim 10^5 \) individual frequency components, and the simultaneous downconversion of all these frequencies in a nonlinear crystal initiates a network of frequency correlations that extends across the width of the resultant comb. The laser source utilized to create the entangled comb is a titanium:sapphire modelocked oscillator that delivers \( \sim 6 \)nm FWHM pulses (\( \sim 140 \)fs) centered at 795nm with a repetition rate of 76MHz. This pulse train is frequency-doubled, which serves to pump a below-threshold optical parametric oscillator (OPO) containing a 2mm BIBO
crystal [36]. The state exiting the OPO is analyzed with homodyne detection, in which the spectral composition of the local oscillator (LO) is modified with an ultrafast pulse shaper capable of independent amplitude and phase modulation [37].

The LO spectrum is partitioned in either 4, 6 or 10 bands of equal energy. By scanning the relative phase between the downconverted comb and the LO, the \( x \) and \( p \) quadrature noises are measured of the state projected onto the spectral composition of the LO mode. The quadrature noises are then recorded for each spectral region as well as all possible pairs of regions. Upon doing so, a covariance matrix is assembled that furnishes a full description of the quantum state. Cross correlations of the form \( \langle \hat{x} \hat{p} \rangle \) are observed to be negligible, which enables the covariance matrix to be expressed in a block diagonal form, i.e., one block for the \( x \)-quadrature and another for the \( p \)-quadrature [36], cf. Eq. (4). The obtained noise values of the leading mode for the 4-, 6-, and 10-dimensional comb are shown below. The significant entanglement with respect to the central wings (elements \( \{1, 4\} \)) from the spectral center (elements \( \{2, 3\} \)) results in the genetic algorithm being unable to induce entanglement from an originally separable operation, and therefore is unable to induce entanglement from an originally separable state.

VI. RESULTS

The results of our methodology for the 4-mode states are detailed in the matrix \( \Sigma_{N=4} \) shown below. The significances \( \Sigma \) are calculated according to Eq. (7), and a particular element in the displayed matrix corresponds to the state partition shown at the same position in Eq. (9).

\[
\Sigma_{N=4} = \begin{pmatrix}
0.0108 & -21.6068 \\
-11.2054 & -24.3358 & -24.5905 \\
-13.1712 & -23.5214 & -23.9689 \\
-4.6575 & -20.9264 & -21.6324 \\
-13.1629 & -24.0338 & -24.3247 & -24.6110
\end{pmatrix}
\]

The first entry in the matrix is the trivial partition with only one party, \( \mathcal{I}_1 = \{1, \ldots, N\} \), and, therefore, must not exhibit entanglement. The following fourteen partitions, however, are each entangled to a significant degree (\( |\Sigma| \gtrapprox 3 \)). The partition displaying the highest entanglement significance is the first to become entangled during downconversion, and therefore exhibits the most significant entanglement. Conversely, the least significantly entangled partition corresponds to detaching the spectral wings (elements \( \{1, 4\} \)) from the spectral center (elements \( \{2, 3\} \)). This partition indicates an asymmetric distribution of entanglement with respect to the central frequency of the comb. In general, symmetric quan-
quantum correlations in the comb are stronger since the preponderance of the downconversion events originate from the pump spectral center. Asymmetric frequency correlations originate from downconversion events displaced from the pump central frequency, which therefore occur with lower probability. Due to the fact that the partition \(\{1, 4\} : \{2, 3\}\) demands the highest degree of asymmetric correlations, it possesses a lowered entanglement significance. Nevertheless, the fact that all of the nontrivial partitions are entangled implies that each resolvable frequency band is entangled with every other band (i.e., the complete entanglement of the comb). Importantly, this characteristic of the quantum comb would go unnoticed without the use of entanglement criteria capable of probing higher-order state partitions, i.e., \(K > 2\).

In the case of 6-modes, 203 unique state partitions are possible, and the resultant entanglement metric \(\Sigma\) is displayed in Fig. 1. The results for the entire set of unique partitions of the 10-mode scenario are likewise depicted in Fig. 2. All of the partitions in both the 6- and 10-mode combs are demonstrated to be entangled except for the trivial partition, \(K = 1\), and cannot be entangled.

Specific \(K\)-partitions and their corresponding entanglement metrics \(\Sigma\) are shown in Table II for the ten mode comb state. Within the \(K = 2\) subgroup, the most significantly entangled partition results from bisecting the spectrum at its center, whereas the least significant entangled structure originates from disconnecting the two extreme spectral zones from the remaining spectrum. This result is consistent with previous observations \([23]\) as well as the results shown above for the 4-mode state (i.e., the partition exhibiting the least entanglement significance requires a high degree of asymmetric frequency correlations). Additionally, 41863 partitions (\(\sim 36\%)\) of the 10-mode state reveal an entanglement more significant than that detected for any of the 511 possible state bi-partitions. Hence, a richer understanding of the quantum phenomena implicit in the multimode state is afforded only upon examination of these higher-order state partitions. As before, the complete dissolusion of the frequency comb structure into ten discrete bins is among the most significantly entangled partitions.

\section*{VII. CONCLUSIONS}

In conclusion, we implemented covariance-based, high-order entanglement criteria on the multimode squeezed states contained within an ultrafast frequency comb. A genetic algorithm was exploited to maximize the statistical significance of the determined entanglement. Upon doing so, the criterion identifies entanglement in all of the 14, 202, and 115974 nontrivial partitions of the 4-, 6-, and 10-mode scenarios, respectively. Consequently, the quantum comb exhibits full multipartite entanglement. Importantly, the currently employed criterion was able to identify entanglement not recognizable with traditional separability metrics. To the best of the authors’ knowledge, the present approach is the first complete entanglement test of a highly complex quantum state.

\section*{Acknowledgement}

This work has been supported by Deutsche Forschungsgemeinschaft through SFB 652, the European Research Council starting grant Freecum, and the French National Research Agency project Comb. C.F. is a member of the Institut Universitaire de France. J.R. acknowledges support from the European Commission through Marie Curie Actions.
FIG. 2. The verified entanglement for all 115974 nontrivial partitions – sorted by significance $\Sigma$ – for the 10-mode frequency-comb Gaussian state.

[1] A. Einstein, N. Rosen, and B. Podolsky, Phys. Rev. 47, 777 (1935).
[2] M. A. Nielsen and I. L. Chuang, Quantum Computation and Quantum Information (Cambridge University Press, Cambridge, England, 2000).
[3] R. Horodecki, P. Horodecki, M. Horodecki, and K. Horodecki, Rev. Mod. Phys. 81, 865 (2009).
[4] O. Gühne and G. Tóth, Phys. Rep. 474, 1 (2009).
[5] M. Huber and J. I. de Vicente, Phys. Rev. Lett. 110, 030501 (2013).
[6] F. Levi and F. Mintert, Phys. Rev. Lett. 110, 150402 (2013).
[7] G. Giedke and B. Kraus, Phys. Rev. A 89, 052303 (2014).
[8] A. Peres, Phys. Rev. Lett. 77, 1413 (1996).
[9] R. Simon, Phys. Rev. Lett. 84, 2726 (2000).
[10] L.-M. Duan, G. Giedke, J. I. Cirac, and P. Zoller, Phys. Rev. Lett. 84, 2722 (2000).
[11] R. F. Werner and M. M. Wolf, Phys. Rev. Lett. 86, 3658 (2001).
[12] J. DiGuglielmo, A. Sambolskiw, B. Hage, C. Pineda, J. Eisert, and R. Schnabel, Phys. Rev. Lett. 107, 240503 (2011).
[13] P. van Loock and A. Furusawa, Phys. Rev. A 67, 052315 (2003).
[14] G. S. Agarwal and A. Biswas, New J. Phys. 7, 211 (2005).
[15] E. Shchukin and V. Vogel, Phys. Rev. Lett. 95, 230502 (2005).
[16] A. Serafini, Phys. Rev. Lett. 96, 110402 (2006).
[17] P. Hyllus and J. Eisert, New J. Phys. 8, 51 (2006).
[18] E. Shchukin and W. Vogel, Phys. Rev. A 74, 030302(R) (2006).
[19] M. Hillery and M. S. Zubaery, Phys. Rev. Lett. 96, 050503 (2006).
[20] O. Gühne, P. Hyllus, O. Gittsovich, and J. Eisert, Phys. Rev. Lett. 99, 130504 (2007).
[21] A. Miranowicz, M. Piani, P. Horodecki, and R. Horodecki, Phys. Rev. A 80, 052303 (2009).
[22] F. Shahandeh, J. Sperling, and W. Vogel, Phys. Rev. A 88, 062323 (2013).
[23] X. Su, A. Tan, X. Jia, J. Zhang, C. Xie, and K. Peng, Phys. Rev. Lett. 98, 070502 (2007).
[24] M. Yukawa, R. Ukai, P. van Loock, and A. Furusawa, Phys. Rev. A 78, 012301 (2008).
[25] M. Pysher, Y. Miwa, R. Shahrokhshahi, R. Bloomer, and O. Pfister, Phys. Rev. Lett. 107, 030505 (2011).
[26] S. Armstrong, J.-F. Morizur, J. Janousek, B. Hage, N. Treps, P. K. Lam, and H.-A. Bachor, Nat. Commun. 3, 1026 (2012).
[27] A. S. Coelho, F. A. S. Barbosa, K. N. Cassemiro, A. S. Villar, M. Martinelli, and P. Nussenzveig, Science 326, 823 (2009).
[28] C. E. Vollmer, D. Schulze, T. Eberle, V. Händchen, J. Fiurášek, and R. Schnabel, Phys. Rev. Lett. 111, 230505 (2013).
[29] M. Horodecki, P. Horodecki, and R. Horodecki, Phys. Lett. A 233, 1 (1996).
[30] M. Horodecki, P. Horodecki, and R. Horodecki, Phys. Lett. A 283, 1 (2001).
[31] J. Sperling and W. Vogel, Phys. Rev. Lett. 111, 110503 (2013).
[32] J. Sperling and W. Vogel, Phys. Rev. A 79, 022318 (2009).
[33] J. Roslund, R. Medeiros de Arájo, S. Jiang, C. Fabre, and N. Treps, Nature Photon. 8, 109 (2014).
[34] G. Adesso and F. Illuminati, J. Phys. A: Math. Theor 40, 7821 (2007).
[35] D. Berend and T. Tassa, Probability and Mathematical Statistics 30, 185 (2010).
[36] R. M. de Araújo, J. Roslund, Y. Cai, G. Ferrini, C. Fabre, and N. Treps, Phys. Rev. A 89, 053828 (2014).
[37] A. M. Weiner, Rev. Sci. Instrum. 71, 1929 (2000).