Uncovering Quantum Correlations with Time Multiplexed Click Detection

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We report on the implementation of a time-multiplexed click detection scheme to probe quantum correlations between different spatial optical modes. We demonstrate that such measurement setups can uncover nonclassical correlations in multimode light fields even if the single mode reductions are purely classical. The nonclassical character of correlated photon pairs, generated by a parametric down-conversion, is immediately measurable employing the theory of click counting instead of low intensity approximations with photoelectric detection models. The analysis is based on second and higher-order factorial moments, which are directly accounted from the measured click statistics. No data post-processing is required to demonstrate the effects of interest with high significance, despite low efficiencies and experimental imperfections. Our approach shows that such novel detection schemes are a reliable and robust way to characterize quantum correlated light fields for practical applications in quantum communications.

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I. INTRODUCTION

Certifying quantum features of light is one key requirement for optical implementations of quantum information technology [1, 2]. This requires, on the one hand, reliable sources for correlated quantum light and, on the other hand, appropriate detection schemes [3, 4]. Since correlations between different degrees of freedom can have different origins in quantum optics, they might be covered by classical statistical optics, or they are genuine quantum properties having no such classical counterpart. Using the classical theory of coherence, one way to discern quantum from classical effects has been introduced independently by R. J. Glauber [5] and E. C. G. Sudarshan [6].

A quantum state characterization is typically based on the photon number distribution; see, e.g., [7, 8]. The corresponding photoelectric detection theory yields Poissonian statistics for coherent light. However, detectors which directly measure photon numbers are typically not available or require advanced data post-processing [9, 10]. Today, quantum states with low mean photon number are often detected with avalanche photodiodes (APDs) in Geiger mode, which basically produce a “click” if any number of photons is absorbed, and remain silent otherwise. A uniform splitting of a radiation field with many photons into portions of lower intensities, each being measured with an APD, can extent the knowledge about the signal, for example, to discriminate single and two-photon events. Various kinds of such photon-number-resolving detectors have been implemented to demonstrate nonclassical features of radiation fields, e.g., in [11-17], or for characterizing the non-Poissonian behavior of the click statistics [18, 19]. One particular realization of such a scheme that requires only a small number of optical elements are so-called time-bin multiplexing detectors (TMD) [18, 20, 21], cf. Fig. 1. Based on these TMD detectors, one can reconstruct nonclassical features of quantum light fields [22, 23] or higher-order correlation functions [24].

The proper theoretical detection model for such devices is a quantum version of the binomial statistics [25]. It was also shown that approximating these statistics with a Poissonian distribution and applying quantumness probes of the photon statistics can yield fake signatures of nonclassicality for classical fields – even if the number of registered signal photons is one order of magnitude below the number of available APDs. To eliminate these errors, nonclassicality criteria in terms of moments of the click counting statistics have been proposed to directly uncover quantum light with click counters without

FIG. 1: (Color online) The realization of two TMDs in a single device is depicted. One part (upper, green input) of the correlated signal field is delayed in an optical fiber before entering the device. Within the TMD, any signal is split by a 50/50 beam splitter followed by another delay line in one output. This multiplexing is done three times yielding 2^3 separated time bins for each (upper green and lower blue) input field which are measured with two APDs.
data post-processing [26]. For example, the notion of sub-binomial light has been introduced [27] and experimentally demonstrated [28, 29].

In the present work, we report on the characterization of a bipartite quantum correlated light source using click detectors only. We demonstrate that click detection is capable to directly verify nonclassicality even for imperfect experimental settings. For our parametric down-conversion (PDC) based light source, this approach correctly shows classical single-mode correlations and, at the same time, it uncovers quantum correlations between the modes. Moreover, we show that our simple treatment to infer these quantum features works for a broad range of pump powers. In particular, it works increasingly well for increasing pump powers, where the often used weak signal approximations with the standard photoelectric detection theory completely break down, as we will show later.

II. NONCLASSICAL MOMENTS OF THE CLICK STATISTICS

The probability to measure \( k_A \) clicks within the \( N = 8 \) time-bins assigned to the signal \( A \) together with \( k_B \) clicks from the signal \( B \), cf. Fig. 1 is described through the joint click-counting statistics \([25, 26]\):

\[
c_{k_A, k_B} = \langle \hat{n}_A^N \hat{m}_A^{N-k_A} (\hat{1}_A - \hat{m}_A)^{k_A} \rangle \times \langle \hat{n}_B^M \hat{m}_B^{M-k_B} (\hat{1}_B - \hat{m}_B)^{k_B} \rangle,
\]

with \( \hat{m}_i = \hat{n}_i - \nu \) denoting the normally ordering prescription and \( \hat{n}_i = e^{-\Gamma(n_i)} \) for the modes \( i = A, B \). In general, \( \Gamma \) can be an unknown detector response being a function of the photon number operators \( \hat{n}_i \). For example, a linear form of the response function is \( \Gamma(\hat{n}_i) = \eta \hat{n}_i / N + \nu \), with \( \eta \) and \( \nu \) being the quantum efficiency and the dark count rate, respectively.

In Ref. [29] it has been demonstrated that the matrix of click moments \( M^{(K_A, K_B)} \) is non-negative for any classical light field,

\[
0 \leq M^{(K_A, K_B)} = \langle \hat{m}_A^{s_A} \hat{m}_B^{s_B} \rangle \langle \hat{n}_A^{t_A} \hat{n}_B^{t_B} \rangle_{(s_A, s_B), (t_A, t_B)},
\]

with \( s_i, t_i = 0, \ldots, K_i/2 \leq N/2 \) for even \( K_i \) and \( N \). For instance, the single-mode and bipartite, second-order matrices of click moments are

\[
M^{(2,0)} = \left( \begin{array}{cc} 1 & \langle \hat{m}_A^2 \rangle \\ \langle \hat{m}_A \rangle & 1 \end{array} \right),
\]

\[
M^{(0,2)} = \left( \begin{array}{cc} 1 & \langle \hat{m}_B^2 \rangle \\ \langle \hat{m}_B \rangle & 1 \end{array} \right),
\]

and

\[
M^{(2,2)} = \left( \begin{array}{ccc} 1 & \langle \hat{m}_A \rangle & \langle \hat{m}_A^2 \rangle \\ \langle \hat{m}_A \rangle & 1 & \langle \hat{m}_A \hat{m}_B \rangle \\ \langle \hat{m}_A \hat{m}_B \rangle & \langle \hat{m}_A \hat{m}_B \rangle & 1 \end{array} \right),
\]

respectively. In the sense of [2], a matrix is classical if all eigenvalues are non-negative, or non-classical if at least one eigenvalue is negative. The needed moments can be directly retrieved from the measured click counting statistics [26].

\[
\langle \hat{m}_A^{i_A} \hat{m}_B^{i_B} \rangle = \sum_{k_A=0}^{N-k_A} \sum_{k_B=0}^{N-k_B} \langle i_A \rangle \langle i_B \rangle c_{k_A, k_B}.
\]

One way to probe the character of nonclassical correlations, i.e. violating inequality [2], can be done as follows. We have nonclassical \( K \)-th order click correlations if

\[
M^{(K,0)} \geq 0, \; M^{(0,K)} \geq 0, \; \text{and} \; M^{(K,K)} \neq 0.
\]

This means that both \( K \)-th order single-mode marginals are classical and the bimodal, \( K \)-th order correlation matrix is nonclassical. In order to genuinely certify such nonclassical correlations, it is sufficient to show that the minimal eigenvalues of the single-mode click-moment matrices \( M^{(K,0)} \) and \( M^{(0,K)} \) are non-negative, and, at the same time, the minimal eigenvalue of the matrix \( M^{(K,K)} \) of joint click moments is negative. Say \( f_A, f_B, \) and \( f_{AB} \) are the normalized eigenvectors to the minimal eigenvalues \( e_A, e_B, \) and \( e_{AB} \) of \( M^{(K,0)} \), \( M^{(0,K)} \) and \( M^{(K,K)} \), respectively. Now, definition [5] is rewritten as

\[
e_A = f_A^\dagger M^{(K,0)} f_A \geq 0, \; e_B = f_B^\dagger M^{(0,K)} f_B \geq 0, \quad \text{and} \quad e_{AB} = f_{AB}^\dagger M^{(K,K)} f_{AB} < 0.
\]

This method will serve as our approach to determine \( K \)-th order quantum correlations between the subsystems \( A \) and \( B \); see the Appendix for further details.

III. IMPLEMENTATION AND MODEL

The states which we investigate for nonclassical correlations are produced in a type II PDC in a periodically poled, 8 mm long KTP waveguide. The PDC process is pumped with 1 ps long pulses at a repetition rate of 70 kHz and a wavelength of 768 nm coming from a Ti:Sapphire laser. The states are generated in two orthogonally polarized signal and idler modes at 1536 nm. The states are produced in a type II PDC in a periodically poled, 8 mm long KTP waveguide. The states are generated in two orthogonally polarized signal and idler modes at 1536 nm. The states are generated in two orthogonally polarized signal and idler modes at 1536 nm.
The PDC may be formulated in terms of the effective Hamiltonian \( \hat{H} = i\kappa \hat{a} \hat{b}^\dagger + H.c., \) where \( \kappa \) is a coupling constant, \( \gamma \) is the coherent amplitude of the pump beam, and \( \hat{a} \) (\( \hat{b} \)) is the creation operator of the mode \( A(B) \). An idealized, perfect unitary evolution for this process yields the two-mode squeezed-vacuum state,

\[
|\xi\rangle = e^{\xi (\hat{a} \hat{b} - \hat{b} \hat{a})} |\text{vac}\rangle \sum_{n=0}^{\infty} \frac{(\tanh \xi)^n}{\cosh \xi} |n\rangle_A |n\rangle_B,
\]

where \( \xi \geq 0 \) is proportional to the square root of the pump power \( P \), since \( \hat{H} \propto \gamma \) and \( P \propto \gamma^2 \). Due to the pairwise generation of photons, this state has perfect photon-number correlations. This means that whenever a certain number of photon is present in one mode, the same number of photons occurs in the other mode. However, the click counting statistics include off-diagonal elements, \( c_{kA,kB} \neq 0 \) for \( k_A \neq k_B \), even for a perfect detection without dark counts and unit efficiency, cf. Fig. 2. This difference between click-counting and photoelectric detection is due to a finite probability that more than one photon can end in the same time-bin. It is worth mentioning that the single-mode reduced states, \( \text{tr}_A[|\xi\rangle\langle \xi|] \) and \( \text{tr}_B[|\xi\rangle\langle \xi|] \), are classical thermal states, and that the total number of photons is \( \langle \hat{n}_A + \hat{n}_B |\xi\rangle = 2 \sinh^2 \xi \).

IV. SECOND-ORDER CORRELATIONS

For the time being, let us focus on the analysis of second-order click correlations, cf. the matrices of moments in [3]. In Fig. 3 we plot the measurement results. Using the approach in [6], the minimal eigenvalues \( e_{AB} \) (top), \( e_A \) (bottom, left), and \( e_B \) (bottom, right) are shown in dependence of the energy per pulse, \( E_{\text{pump}} = P/70 \text{kHz} \), for the second-order matrices \( M^{(2,2)} \), \( M^{(2,0)} \), and \( M^{(0,2)} \), respectively. The single-mode matrices are non-negative, \( e_A \geq 0 \) and \( e_B \geq 0 \), whereas the bipartite correlations are nonclassical, \( e_{AB} < 0 \). Thus, we successfully verified the quantum nature of the second-order click correlations between the spatial modes \( A \) and \( B \).

In order to compare our measured results, a simple theoretical model is used. We assume that the pure state \( |\xi\rangle \) is generated, and the detectors are described via a plain linear response function: \( \Gamma(\hat{n}/N) = \eta \hat{n}/N + \nu \). As we discussed above, the parameter \( \xi \), characterizing the state \( |\xi\rangle \), depends on the pump power \( P \): \( \xi = \xi_0 \sqrt{P} \). The proportionality constant \( \xi_0 \), the quantum efficiency \( \eta \), and the dark count rate have been fitted using the standard method of least squares. They are \( \eta = 9.6\% \), \( \nu = 0.51 \), and \( \xi_0 = 0.087 \text{ (\mu W)^{-1/2}} \). With these values, we estimate total mean photon numbers, of the model state \( |\xi\rangle \) in Eq. (7), to be 0.9...15 photons for the pump powers 50...403 \( \text{\mu W} \) or energies 0.7...5.8 nJ. Already this simplified model yields a good agreement with the measured results, which highlights the excellent performance of the engineered PDC source; cf. the dashed lines in Fig. 3.

The inset in the upper plot shows the extrapolation...
of the quantum correlations, $e_{AB} < 0$, for higher pump energies. At some point, the mean intensity is so high, that these correlations saturate and eventually vanish. A high squeezing level and a classical laser light with a large coherent amplitude result in the same signal – all time-bins are occupied with a large number of photons. Therefore, at high mean intensities, the signals of nonclassical and classical states cannot be discriminated. Thus, the recorded quantum correlations of the former state must decrease due to the properties of click counting devices, which is automatically included in the click counting theory.

Let us emphasize again that the states have been generated for pump powers ranging over almost one order of magnitude, 50 to 403 μW, with a relative error estimate of 10%. Verifying quantum correlations in this comparably large domain is typically considered a challenging task, but can easily be accomplished with our TMD click counters. In addition, since the method of nonclassical click moments is independent of the state, the verified quantum correlations can be certified even if the pump power was completely unknown.

V. HIGHER-ORDER CORRELATIONS

Beyond the considerations of physically meaningful second-order correlations, let us study the verification of higher-order quantum correlations. The highest possible number of correlations one can infer from $N = 8$ time bins for each mode is given by $K = 8$ in Eq. (8). The bound for a classical signal, $M^{(k,K)} \geq 0$, is given by the eigenvalue $e_{cl} = 0$, which can be directly computed for a two-mode coherent state; see also [26]. Thus, the signed distance, in units of standard deviations, of the experimentally obtained minimal eigenvalue, $e = \bar{e} \pm \Delta e$, to this classical bound leads to a signed significance,

$$\Sigma = \frac{\bar{e} - e_{cl}}{\Delta e} = \frac{\bar{e}}{\Delta e}. \quad (8)$$

A negative significance, $\Sigma < 0$, verifies the nonclassicality with a significance of $|\Sigma|$. Typically, $\Sigma \lesssim -3$ is a significant verification of the negativity, whereas $\Sigma \approx 0$ cannot be distinguished from the classical bound 0, i.e., an insignificant result.

In Fig. 4, the significance levels of the eighth order quantum correlation are given. The single-mode, signed significances for $M^{(8,0)}$ and $M^{(0,8)}$ can be additionally found in the Appendix. There are no significant eighth order, single-mode correlations, $M^{(8,0)}, M^{(0,8)} \geq 0$ – the largest single-mode negativities are in the order of $\Sigma \sim -10^{-5}$. Additionally, the significance of negativities in $M^{(8,8)}$, cf. Fig. 4, are in the range of 2.7...10.6 for the pump powers 50...403 μW, respectively. Hence, for most of the pump powers, a significant eighth-order nonclassical correlations between the modes $A$ and $B$ is certified via higher order moments. Remarkably, the significance even increases with increasing pump power. On the one hand, one would typically not use such comparably high intensities in our measurement setup, because the difference between photo-counting theory and click-counting is quite pronounced in this regime, cf. Fig. 2. This can also be seen when analyzing the data on purpose with the inappropriate photoelectric detection model. Then, the single-mode signed significances evaluate to $-2.4...-13$, cf. the Appendix, suggesting fake nonclassicality which worsens with increasing pump power. Our consistent treatment in terms of the click statistics, on the other hand, correctly identifies this higher-order bimodal correlations while showing, as expected for our source, no nonclassicality in the single-mode marginals.

VI. CONCLUSIONS

In summary, we set up a click correlation measurement scheme using time-multiplexed detectors for probing nonclassical correlations of a bipartite quantum light field. We could directly infer nonclassical correlations from the measured joint click-counting statistics with high significances. This was possible despite a low detection efficiency, estimated as 10%, and over one order of magnitude of intensities. Note that the bare click data have been analyzed without corrections for eficiencies, dark counts, or related post-processing techniques. We compared our results with a simple theoretical model and obtained a very good agreement. In particular, the expectation of this model – all orders of single-mode correlations do not exhibit nonclassicality, but the two-mode correlations do – is correctly demonstrated with our method. This underlines the functionality of our device, and it simultaneously verifies genuine quantum correlations between the spatial optical modes having single-mode marginals in the form of classical thermal light.

We conclude that our proposed measurement together with the click-detection analysis is a robust and efficient tool to characterize quantum light, and believe that this
simplicity of click detection – being solely a collection of probabilities of coincidence clicks – paves the way towards real-live implementations of quantum communications protocols in optical systems. The present approach may be further generalized to handle more complex types of quantum-correlated, multi-mode radiation fields.

Note that during the completion of this manuscript, we became aware of a related paper in preparation by the group of I. A. Walmsley, University of Oxford.

Appendix A: Measuring the click statistics

All we need is a plain device producing a binary information referred to as “click” and “no-click” or “on/off” events. This can be formally written as the expectations value of a so-called detector response function \( \Gamma(\hat{n}) \), where \( \hat{n} \) is the photon number operator. Thus the positive operator-valued measure is given by the elements

\[
\hat{\Pi}_{\text{click}} = \hat{1} - e^{-\Gamma(\hat{n})}, \quad \text{and} \quad \hat{\Pi}_{\text{no-click}} = e^{-\Gamma(\hat{n})}.
\]

(A1)

Splitting a signal into \( N = 8 \) modes of equal intensities each of them measured with this binary event device, yields the click counting statistics of two detectors [26]:

\[
c_{k_A,k_B} = \langle \hat{N}_{k_A} \rangle = e^{-\Gamma(\hat{n}_A/N)} \left( \hat{1} - e^{-\Gamma(\hat{n}_A/N)} \right)^{N-k_A} \left( \hat{e}^{-\Gamma(\hat{n}_B/N)} \right)^{k_B} \left( \hat{1} - e^{-\Gamma(\hat{n}_B/N)} \right)^{N-k_B},
\]

where \( k_i (i = A, B) \) denote the numbers of coincidence clicks from both detectors. Note that we assume that both detectors \( A \) and \( B \) have the same characteristics, as it is true for our implementation. A typical example is a linear response function, \( \Gamma(\hat{n}/N) = \eta \hat{n}/N + \nu \), with \( \eta \) being the quantum efficiency, and \( \nu \) the dark count rate.

It is worth to stress that \( c_{k_A,k_B} \) is the probability that we have \( k_A \) clicks from the first detector system simultaneous with \( k_B \) clicks of the second one. The experimentally recorded number of coincidence events \((k_A,k_B)\) may be labeled as \( C_{k_A,k_B} \). Hence, we get the relative frequencies

\[
c_{k_A,k_B}^{\exp} = C_{k_A,k_B} \cdot \frac{C}{\sum_{k_A=0}^{N} \sum_{k_B=0}^{N} C_{k_A,k_B}},
\]

(A3)

denoting the total number of events. Later on, the single mode marginals will have some importance. They are given by \( c_{k_A}^{\exp} = \sum_{k_B=0}^{N} c_{k_A,k_B}^{\exp} \) and \( c_{k_B}^{\exp} = \sum_{k_A=0}^{N} c_{k_A,k_B}^{\exp} \).

Appendix B: Click moments from measured click statistics

Let us define the operators

\[
\hat{m}_A^l \hat{m}_B^l := e^{-l_A \Gamma(\hat{n}_A/N)} e^{-l_B \Gamma(\hat{n}_B/N)}.
\]

(B1)

for \( 0 \leq l_i \leq N \ (i = A, B) \). Note that we use here the moments \( \hat{m}_i \) instead of \( \hat{\pi}_i = \hat{1} - \hat{m}_i \), which were used in our earlier work [26]. The mathematical treatment in using \( \hat{m}_i \) or \( \hat{\pi}_i \) is identical. However, it is more convenient applying \( \hat{m}_i \) for the present considerations. The matrix of joint click counting moments is

\[
M^{(N,N)} = \begin{pmatrix}
1 & \langle \hat{m}_A \rangle & \langle \hat{m}_B \rangle & \langle \hat{m}_A^2 \rangle & \langle \hat{m}_A \hat{m}_B \rangle & \langle \hat{m}_A^3 \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \cdots \\
\langle \hat{m}_A \rangle & \langle \hat{m}_A^2 \rangle & \langle \hat{m}_A \hat{m}_B \rangle & \langle \hat{m}_A^3 \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \langle \hat{m}_A^4 \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \cdots \\
\langle \hat{m}_B \rangle & \langle \hat{m}_A \hat{m}_B \rangle & \langle \hat{m}_B^2 \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \langle \hat{m}_A^4 \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \langle \hat{m}_A^2 \hat{m}_B^2 \rangle & \cdots \\
\langle \hat{m}_A^2 \rangle & \langle \hat{m}_A^3 \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \langle \hat{m}_A^4 \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \langle \hat{m}_A^5 \rangle & \langle \hat{m}_A^4 \hat{m}_B \rangle & \cdots \\
\langle \hat{m}_A \hat{m}_B \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \langle \hat{m}_A^4 \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \langle \hat{m}_A^5 \rangle & \langle \hat{m}_A^4 \hat{m}_B \rangle & \cdots \\
\langle \hat{m}_A^2 \rangle & \langle \hat{m}_A^3 \rangle & \langle \hat{m}_A^2 \hat{m}_B \rangle & \langle \hat{m}_A^4 \rangle & \langle \hat{m}_A^3 \hat{m}_B \rangle & \langle \hat{m}_A^5 \rangle & \langle \hat{m}_A^4 \hat{m}_B \rangle & \cdots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix},
\]

(B2)
where the framed part includes the second order correlations \( M^{(2,2)} \). It is denoted as the cross-correlation matrix, since \( \det M^{(2,2)} = \langle [\Delta \hat{m}_A]^2 \rangle \langle [\Delta \hat{m}_B]^2 \rangle - \langle [\Delta \hat{m}_A][\Delta \hat{m}_B] \rangle^2 \). The single-mode reduced matrices of moments read as

\[
M^{(N,0)} = \begin{pmatrix}
1 & \langle \hat{m}_A \rangle & \langle \hat{m}_A^2 \rangle & \cdots \\
\langle \hat{m}_A \rangle & \langle \hat{m}_A^2 \rangle & \langle \hat{m}_A^2 \rangle & \cdots \\
\langle \hat{m}_A^2 \rangle & \langle \hat{m}_A^2 \rangle & \langle \hat{m}_A^2 \rangle & \cdots \\
\vdots & \vdots & \vdots & \ddots
\end{pmatrix}
\quad \text{and} \quad
M^{(0,N)} = \begin{pmatrix}
1 & \langle \hat{m}_B \rangle & \langle \hat{m}_B^2 \rangle & \cdots \\
\langle \hat{m}_B \rangle & \langle \hat{m}_B^2 \rangle & \langle \hat{m}_B^2 \rangle & \cdots \\
\langle \hat{m}_B^2 \rangle & \langle \hat{m}_B^2 \rangle & \langle \hat{m}_B^2 \rangle & \cdots \\
\vdots & \vdots & \vdots & \ddots
\end{pmatrix}
\]

(B3)

Again, the framed inset includes solely the moments up to the second order, i.e.: \( M^{(2,0)} \) and \( M^{(0,2)} \). Those are defined as the variance matrices, because \( \det M^{(2,0)} = \langle [\Delta \hat{m}_A]^2 \rangle \) and \( \det M^{(0,2)} = \langle [\Delta \hat{m}_B]^2 \rangle \).

The moments may be retrieved via [20]

\[
\langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle = \sum_{k_A=0}^{N-l_A} \sum_{k_B=0}^{N-l_B} \frac{(N-k_A)}{l_A!} \frac{(N-k_B)}{l_B!} c_{k_A,k_B},
\]

yielding an experimental mean value of

\[
\frac{\langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle}{C} = \sum_{k_A=0}^{N-l_A} \sum_{k_B=0}^{N-l_B} c_{k_A,k_B} \frac{(N-k_A)}{l_A!} \frac{(N-k_B)}{l_B!}.
\]

(B4)

The standard estimate for the sampling error is

\[
\sigma \left( \frac{\langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle}{\sqrt{C}} \right) = \sqrt{\frac{1}{(C-1)}} \sum_{k_A=0}^{N-l_A} \sum_{k_B=0}^{N-l_B} c_{k_A,k_B} \left( \frac{(N-k_A)}{l_A!} \frac{(N-k_B)}{l_B!} - \frac{\langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle}{\sqrt{C}} \right)^2.
\]

(B5)

For \( 0 \leq K_A, K_B \leq N \), the matrix of sampled mean values, \( \overline{M}(K_A,K_B) \), and the matrix of the errors, \( \Delta M(K_A,K_B) \), are given by the elements \( \langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle \) and \( \sigma \left( \frac{\langle \hat{m}_A^{l_A} \hat{m}_B^{l_B} \rangle}{\sqrt{C}} \right) \), respectively, arranged according to Eqs. [B2] and [B3].

**Appendix C: Determining correlations**

For fully classical states, the matrices of moments are non-negative. There are genuine quantum correlations of \( K \)-th order between the modes, if

\[
M^{(K,0)} \geq 0, \quad M^{(0,K)} \geq 0, \quad \text{and} \quad M^{(K,K)} \geq 0,
\]

for \( 0 \leq K \leq N \). This would mean that there are no non-classical correlation in the single mode marginals, but there are some nonclassical two-mode correlations. The non-negativity of a matrix \( M \), or its violation, may be probed by its normalized eigenvectors \( \vec{v} \) (\( M\vec{v} = \epsilon \vec{v} \), with \( \vec{v}^\dagger \vec{v} = 1 \) and \( \epsilon \) denoting the corresponding eigenvalue), since

\[
\vec{v}^\dagger M \vec{v} = \epsilon.
\]

(C2)

If all eigenvalues of any matrix \( M \) are non-negative, \( \forall \epsilon : \epsilon_n \geq 0 \), it holds that \( M \geq 0 \). Conversely, if at least one eigenvalue is negative, \( \exists \epsilon : \epsilon_n < 0 \), we have a violation of the non-negativity \( M \not\geq 0 \).

Experimentally, this may be obtained via the following steps:

1. determine the matrix of moments, i.e.: \( \overline{M} \pm \Delta M \);
2. compute the normalized eigenvectors \( \vec{v}_n \) of \( \overline{M} \) to get

\[
\vec{v}_n = \vec{v}_n^\dagger \overline{M} \vec{v}_n;
\]
3. perform a linear error propagation, which yields

\[
\Delta e_n = |\vec{v}_n^\dagger| \Delta M |\vec{v}_n|,
\]

where \( |\vec{f}| = (|f_1|, |f_2|, \ldots)^T \) for any vector \( \vec{f} \).
4. and, finally, one gets \( e_n = \bar{e}_n \pm \Delta e_n \), or one may compute the signed significance \( \Sigma_n = \bar{e}_n / \Delta e_n \).

Let us stress that the minimal bound for the eigenvalues of classical states is \( e_{cl} = 0 \), cf. [20]. This means that the absolute value of the significance corresponds to the distance of \( e \) to this classical bound in units of error bars, \( |\Sigma| = |e - e_{cl}| / \Delta e \). Additionally, the sign of \( \Sigma \) shows if we are consistent with this bound, \( \Sigma \gtrless 0 \), or clearly violate it, \( \Sigma \lesssim -3 \), meaning that the underlying state is significantly nonclassical. Any significance level smaller than this value cannot be well separated from the classical bound.

**Appendix D: All results of the data analysis**

In this section, we present all information obtained during our data processing. We used the method as described previously or in the main body. In Fig. 5 the measured click statistics are given. In Tables I and II the eigenvalues of the single mode variances and the second order cross-correlations, respectively, are given. Similarly, Tables III and IV include the significance of the eigenvalues of the full matrix of moment for the bipartite and single mode case, respectively.

**FIG. 5:** The measured click counting statistics \( c_{k_A,k_B} \) (for \( k_A,k_B = 0,\ldots,8 \)) are shown at different pump powers \( \{50 \mu W, 100 \mu W, 200 \mu W, 300 \mu W, 403 \mu W \} \). A blank area depicts that no coincidence has been recorded. The total numbers of events are \( \{41750447, 41755519, 37543468, 41728175, 41711631 \} \), being sorted according to the previous pump powers. The \( c_{0,0} \) coincidences are cut. They have the values \( \{0.96, 0.91, 0.81, 0.70, 0.55 \} \), respectively, yielding the major contribution to the whole statistics, i.e., more than 50%.

**TABLE I:** The eigenvalues \( (e_j^i, i = 1, 2 \text{ and } j = A, B) \) are shown for the single mode variance matrices \( M^{(2,0)} \) and \( M^{(0,2)} \). The minimal (boldface) eigenvalues are plotted in the main body. All eigenvalues are non-negative.

<table>
<thead>
<tr>
<th>pump power</th>
<th>( e_1^A )</th>
<th>( e_1^B )</th>
<th>( e_2^A )</th>
<th>( e_2^B )</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 ( \mu W )</td>
<td>1.9936867( \times10^{-6} )</td>
<td>1.9941778( \times10^{-6} )</td>
<td>(33.3( \pm )6.5)( \times10^{-6} )</td>
<td>(36.3( \pm )6.3)( \times10^{-6} )</td>
</tr>
<tr>
<td>100 ( \mu W )</td>
<td>1.985126( \times10^{-5} )</td>
<td>1.9863857( \times10^{-6} )</td>
<td>(9.2( \pm )1.0)( \times10^{-5} )</td>
<td>(97.3( \pm )9.8)( \times10^{-6} )</td>
</tr>
<tr>
<td>200 ( \mu W )</td>
<td>1.966550( \times10^{-5} )</td>
<td>1.968691( \times10^{-5} )</td>
<td>(27.0( \pm )1.6)( \times10^{-5} )</td>
<td>(28.2( \pm )1.6)( \times10^{-5} )</td>
</tr>
<tr>
<td>300 ( \mu W )</td>
<td>1.940763( \times10^{-5} )</td>
<td>1.945466( \times10^{-5} )</td>
<td>(64.0( \pm )2.1)( \times10^{-5} )</td>
<td>(63.0( \pm )2.0)( \times10^{-5} )</td>
</tr>
<tr>
<td>403 ( \mu W )</td>
<td>1.894942( \times10^{-5} )</td>
<td>1.897773( \times10^{-5} )</td>
<td>(165.7( \pm )2.9)( \times10^{-5} )</td>
<td>(170.9( \pm )2.9)( \times10^{-5} )</td>
</tr>
</tbody>
</table>
TABLE II: The eigenvalues \( e_i^{AB}, i = 1, 2, 3 \) of the click correlations matrix \( M^{(2,2)} \) are shown. The negative (boldface) eigenvalues are plotted.

\[
\begin{array}{|c|c|c|c|}
\hline
\text{pump power} & e_1^{AB} & e_2^{AB} & e_3^{AB} \\
\hline
50 \mu W & 3.976098 \pm 2.1 \times 10^{-5} & (148.4 \pm 9.8) \times 10^{-6} & (-1.5 \pm 1.1) \times 10^{-5} \\
100 \mu W & 3.944098 \pm 3.2 \times 10^{-5} & (39.3 \pm 1.5) \times 10^{-5} & (-3.6 \pm 1.8) \times 10^{-5} \\
200 \mu W & 3.873915 \pm 5.2 \times 10^{-5} & (111.0 \pm 2.5) \times 10^{-5} & (-9.0 \pm 2.8) \times 10^{-5} \\
300 \mu W & 3.780747 \pm 6.5 \times 10^{-5} & (243.5 \pm 3.2) \times 10^{-5} & (-15.2 \pm 3.6) \times 10^{-5} \\
403 \mu W & 3.608217 \pm 8.8 \times 10^{-5} & (604.6 \pm 4.7) \times 10^{-5} & (-32.2 \pm 4.9) \times 10^{-5} \\
\hline
\end{array}
\]

TABLE III: The signed significances \( \Sigma_i^{AB} \) of eigenvalues \( e_i^{AB}, i = 1, \ldots, 10 \) of the full higher-order click correlations of the \( 10 \times 10 \) bipartite matrix of moments \( M^{(8,8)} \) are given depending on the pump power. The third eigenvalues are negative with the highest significance \( (\Sigma_3 \lesssim -3) \) and, thus, clearly demonstrate the nonclassical character of the generated radiation field.

\[
\begin{array}{|c|c|c|c|c|c|c|c|c|}
\hline
\text{pump power} & \Sigma_1^{AB} & \Sigma_2^{AB} & \Sigma_3^{AB} & \Sigma_4^{AB} & \Sigma_5^{AB} & \Sigma_6^{AB} & \Sigma_7^{AB} & \Sigma_8^{AB} & \Sigma_9^{AB} & \Sigma_{10}^{AB} \\
\hline
50 \mu W & 6.0 \times 10^4 & 2.3 \times 10^1 & -2.7 \times 10^0 & 2.2 \times 10^{-2} & -2.9 \times 10^{-3} & -8.7 \times 10^{-4} & 2.0 \times 10^{-5} & 5.7 \times 10^{-6} & 3.7 \times 10^{-6} & 2.8 \times 10^{-6} \\
100 \mu W & 3.9 \times 10^4 & 3.9 \times 10^1 & -4.0 \times 10^0 & 5.0 \times 10^{-2} & -6.0 \times 10^{-3} & -1.3 \times 10^{-3} & 7.2 \times 10^{-5} & 2.9 \times 10^{-5} & 9.2 \times 10^{-6} & 6.9 \times 10^{-6} \\
200 \mu W & 2.4 \times 10^4 & 6.5 \times 10^1 & -5.9 \times 10^0 & 1.6 \times 10^{-1} & -1.5 \times 10^{-2} & -3.0 \times 10^{-3} & 2.1 \times 10^{-4} & -7.4 \times 10^{-5} & 2.3 \times 10^{-5} & 1.4 \times 10^{-5} \\
300 \mu W & 1.9 \times 10^4 & 1.0 \times 10^2 & -7.5 \times 10^0 & 5.0 \times 10^{-1} & -3.2 \times 10^{-2} & -2.0 \times 10^{-3} & 1.9 \times 10^{-4} & -2.5 \times 10^{-5} & -8.5 \times 10^{-5} & 1.5 \times 10^{-5} \\
403 \mu W & 1.4 \times 10^4 & 1.7 \times 10^2 & -1.1 \times 10^1 & 1.7 \times 10^0 & -9.3 \times 10^{-2} & 1.5 \times 10^{-2} & -1.6 \times 10^{-3} & -9.4 \times 10^{-4} & 4.6 \times 10^{-4} & 1.1 \times 10^{-4} \\
\hline
\end{array}
\]

TABLE IV: The signed significance \( \Sigma_i^j \) of eigenvalues \( e_i^j, i = 1, \ldots, 5 \) and \( j = A, B \) of the full higher-order moments are given for the single-mode reductions \( A \) and \( B \). The single-mode matrices of moments \( M^{(8,0)} \) and \( M^{(0,8)} \) are of the dimensionality \( 5 \times 5 \). All significant values, \( \Sigma \gtrsim 3 \), are non-negative, and most of the insignificant values are positive too. Note that any insignificant negative value, \( \Sigma \lesssim 0 \), is not inconsistent with the classical bound 0 and, thus, it cannot imply nonclassicality.

\[
\begin{array}{|c|c|c|c|c|c|}
\hline
\text{pump power, mode} & \Sigma_1^A & \Sigma_2^A & \Sigma_3^A & \Sigma_4^A & \Sigma_5^A \\
\hline
50 \mu W, j = A & 8.7 \times 10^4 & 1.6 \times 10^1 & 2.3 \times 10^{-4} & -2.4 \times 10^{-5} & -7.7 \times 10^{-12} \\
50 \mu W, j = B & 9.1 \times 10^4 & 1.8 \times 10^1 & 7.5 \times 10^{-5} & -4.9 \times 10^{-5} & -8.4 \times 10^{-10} \\
100 \mu W, j = A & 5.7 \times 10^4 & 2.8 \times 10^1 & 1.4 \times 10^{-3} & -1.6 \times 10^{-5} & 3.0 \times 10^{-10} \\
100 \mu W, j = B & 5.9 \times 10^4 & 3.1 \times 10^1 & 2.7 \times 10^{-3} & 2.4 \times 10^{-5} & -6.3 \times 10^{-8} \\
200 \mu W, j = A & 3.6 \times 10^4 & 5.0 \times 10^1 & 1.6 \times 10^{-2} & -2.1 \times 10^{-5} & -3.6 \times 10^{-8} \\
200 \mu W, j = B & 3.7 \times 10^4 & 5.3 \times 10^1 & 2.0 \times 10^{-2} & 3.9 \times 10^{-5} & -3.5 \times 10^{-7} \\
300 \mu W, j = A & 2.8 \times 10^4 & 8.6 \times 10^1 & 8.8 \times 10^{-2} & 5.5 \times 10^{-5} & -4.1 \times 10^{-6} \\
300 \mu W, j = B & 2.9 \times 10^4 & 8.9 \times 10^1 & 7.7 \times 10^{-2} & 1.4 \times 10^{-4} & -9.2 \times 10^{-7} \\
403 \mu W, j = A & 2.1 \times 10^4 & 1.5 \times 10^2 & 4.0 \times 10^{-1} & 1.2 \times 10^{-3} & -1.9 \times 10^{-7} \\
403 \mu W, j = B & 2.1 \times 10^4 & 1.6 \times 10^2 & 4.0 \times 10^{-1} & 8.8 \times 10^{-4} & 1.4 \times 10^{-6} \\
\hline
\end{array}
\]

Appendix E: Fake effects

The typically considered approximation is that the photoelectric detection model is valid for a large number of bins \( N \) and for low intensities. In other words, one assumes

\[
p_{k_A,k_B} = \langle \Gamma(\hat{n}_A)^{k_A} \rangle_{\hat{k_A}!} e^{-\Gamma(\hat{n}_A)} \langle \Gamma(\hat{n}_B)^{k_B} \rangle_{\hat{k_B}!} e^{-\Gamma(\hat{n}_B)} \approx c_{k_A,k_B}. \tag{E1}
\]

The moments of the photoelectric counting statistics \( p_{k_A,k_B} \) are given by

\[
\langle [\Gamma(\hat{n}_A)]^{m_A} [\Gamma(\hat{n}_B)]^{m_B} \rangle = \sum_{k_A \geq m_A} \sum_{k_B \geq m_B} \frac{k_A!}{(k_A - m_A)!} \frac{k_B!}{(k_B - m_B)!} p_{k_A,k_B}. \tag{E2}
\]

In the simplest case of \( \Gamma(\hat{n}) = \hat{n} \), these moments correspond to the normally ordered photon number moments, \( \langle \hat{n}_A^{m_A} \hat{n}_B^{m_B} \rangle \). In order to prove that fake effects occur, we follow the same treatment as done for the click-counting
moments. That is, we formulate the single mode $5 \times 5$ matrices of photoelectric moments of the marginals, i.e.,

$$\langle \left[ \Gamma(\hat{n}_A) \right]^{m_A+m_A'} \rangle_N^{m_A=0} \quad \text{and} \quad \langle \left[ \Gamma(\hat{n}_B) \right]^{m_B+m_B'} \rangle_N^{m_B=0},$$

and compute the signed significances $\Sigma^A$ and $\Sigma^B$ of the minimal eigenvalue. If the approximation (E1) was true, then the marginal thermal states must not exhibit significant negativities. The corresponding falsification with our data is given in Table V. This simply means that the approximation in Eq. (E1) is not justified for the used detection scheme.

**TABLE V:** The minimal signed significances $\Sigma^j$ ($j = A, B$) of the eighth-order photoelectric moments are given for the single-mode reductions $A$ and $B$. The negativities demonstrate that the photoelectric detection model is not adequate for our detection scheme. They tend to increase with increasing pump powers. Apparently the fake effects are highly sensitive to the modes – $A$ corresponds to the earlier time bins and $B$ to the later ones – which might be due to an overestimation of afterpulses using the wrong measurement statistics.

<table>
<thead>
<tr>
<th>pump power</th>
<th>$\Sigma^A$</th>
<th>$\Sigma^B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50 $\mu$W</td>
<td>$-2.6$</td>
<td>$-2.4$</td>
</tr>
<tr>
<td>100 $\mu$W</td>
<td>$-3.1$</td>
<td>$-1.4$</td>
</tr>
<tr>
<td>200 $\mu$W</td>
<td>$-2.6$</td>
<td>$-2.2$</td>
</tr>
<tr>
<td>300 $\mu$W</td>
<td>$-6.6$</td>
<td>$-3.1$</td>
</tr>
<tr>
<td>403 $\mu$W</td>
<td>$-12$</td>
<td>$-13$</td>
</tr>
</tbody>
</table>


