Faithful hierarchy of genuine $n$-photon quantum non-Gaussian light

Lukáš Lachman, Ivo Straka, Josef Hloušek, Miroslav Ježek, and Radim Filip

Department of Optics, Faculty of Science, Palacký University,
17. listopadu 1192/12, 771 46 Olomouc,
Czech Republic

Light is an essential tool for connections between quantum devices and for diagnostic of processes in quantum technology. Both applications deal with advanced nonclassical states beyond Gaussian coherent and squeezed states. Current development requires a loss-tolerant diagnostic of such nonclassical aspects. We propose and experimentally verify a faithful hierarchy of genuine $n$-photon quantum non-Gaussian light. We conclusively witnessed 3-photon quantum non-Gaussian light in the experiment. Measured data demonstrates a direct applicability of the hierarchy for a large class of real states.

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Individual photons as bosonic elementary particles have been subjects of a detailed quantum analysis already for many decades. It is intensified now due to their importance for quantum technology. First, a single photon antibunching was measured as incompatible with classical coherence theory \[1, 2\]. It was the first test of nonclassical light. This measurement became canonical for single photon sources \[1–5\]. After many years, broadband homodyne detection allowed indirect estimation of their continuous variable nonclassical features \[6–11\]. Their visualisation in a phase space of continuous amplitude of electric field by a Wigner probability density function shows multiple negative concentric annuli for Fock states of light \[12\]. Wigner functions are used to distinguish different Fock states of light, however, without any proof yet that they really form a faithful hierarchy. A faithful hierarchy of $n$-photon quantum non-Gaussian states would reliably recognize that, for a given order $n$, an observed state is statistically incompatible with any mixture of Fock-state superpositions up to $|n-1\rangle$ modified by an arbitrary Gaussian phase-space transformation \[10, 13, 14\]. Unfortunately, such a faithful hierarchy based on the negative parts of Wigner function has not been discovered yet and it would be anyway applicable only if overall losses were below fifty percent. Since a large variety of experimental platforms emitting or transmitting light does not suppress the losses so much, a lack of theoretical tools witnessing genuine $n$-photon quantum non-Gaussianity limits optical diagnostic of quantum processes in matter, current fast development of multiphoton sources and their applications in quantum technology.

A large gap between basic nonclassical light and light with negativity of Wigner function was partially covered when a loss-tolerant direct measurement of single-photon quantum non-Gaussianity was proposed and immediately experimentally tested \[15, 16\]. Advantageously, these criteria use only basic multi-photon correlation measurements, commonly applied to verify nonclassicality. The quantum non-Gaussianity criteria conclusively prove that light is not compatible with any mixture of Gaussian states, even beyond fifty percent of loss \[17\]. In difference to the tests of nonclassicality, such test of quantum non-Gaussianity can already recognize a much narrower set of states, approaching closer to ideal single photon states. That property of single photon states has been already proposed to be applicable as a security indicator of single-photon quantum key distribution \[19\] and as a...

*These authors contributed equally to this work.
parameters $\alpha, \beta$.

In both cases, it was proven that a test of nonclassicality is not sufficient and it can lead to misleading results. Recently, criteria of quantum non-Gaussianity for multiphoton light have been proposed and measured despite very large optical loss [18]. Meanwhile, quantum non-Gaussianity criteria have been developed for other types of the states [1] [21] [22]. Recent mathematical treatment to quantum non-Gaussianity led to a formulation of a resource theory [24] [25].

The extension to multi-photon light allows wider applications in diagnostic of quantum processes, but the criteria [18] do not still form the faithful hierarchy of quantum properties and therefore, such genuine $n$-photon quantum non-Gaussian state can not be directly witnessed under large optical loss. Discovery of the hierarchy is currently crucial for ongoing exploration of light emitted by higher order nonlinear processes [26, 27] and for current development of multiphoton sources [13] [28]. In this Letter, we derived the faithful hierarchy of sufficient conditions for genuine $n$-photon quantum non-Gaussian state and, simultaneously, we experimentally verified the hierarchy by photon counting measurement of light up to three photons under 6.5 dB of optical loss. Under such loss, negative Wigner function cannot be observed. Our criteria can conclusively confirm that observed genuine $n$-photon quantum non-Gaussian statistics is beyond statistics produced by any mixture of superposition of $n-1$ photons possibly modified by any Gaussian transformation. We further analyzed and experimentally verified robustness of the genuine multiphoton state under background noise. All these results qualify this faithful loss-tolerant hierarchy presented here to be both of fundamental interest and also directly applicable in many laboratories.

A pure state $|\psi\rangle$ exhibits genuine $n$-photon quantum non-Gaussianity if it can not be expressed as

$$|\psi\rangle \neq S(\beta)D(\alpha)|\tilde{\psi}_{n-1}\rangle,$$

where the core state $|\tilde{\psi}_{n-1}\rangle$ represents any superposition of Fock states $|0\rangle,...,|n-1\rangle$ that can be affected by displacement $D(\alpha) = \exp(\alpha a^\dagger - \alpha^* a)$ or by squeezing $S(\beta) = \exp[-\beta (a^\dagger)^2 + \beta^* a^2]$ operation. The Gaussian transformation $S(\beta)D(\alpha)$ can add only a Gaussian envelope to the core state $|\tilde{\psi}_{n-1}\rangle = \sum_{m=0}^{n-1} c_m |m\rangle$ [29]. The envelope changes photon statistics, however, it only scales the shape of the Wigner function representing the state in the phase space. Single-mode impure state determined by density matrix $\rho$ shows the $n$th ordered property if it is not identified with any statistical mixture of the right side in inequality (1) over the complex parameters $\alpha$, $\beta$ and $c_m$. That introduces a hierarchy of quantum non-Gaussian attributes labeled by an index $n$. Obviously, each ideal Fock state $|n\rangle$ has an attribute of order $n$ which any lower Fock state does not achieve even after any Gaussian transformation. The lowest (first) order of the hierarchy is quantum non-Gaussianity proposed and measured in Refs. [15] [19]. The second order means that observed photon statistics are not compatible with any mixture of states $S(\beta)D(\alpha)(c_0|0\rangle + c_1|1\rangle)$ for any complex $\alpha$, $\beta$, $c_0$ and $c_1$ satisfying $|c_0|^2 + |c_1|^2 = 1$. In this case, the Gaussian transformation $S(\beta)D(\alpha)$ increases over one maximal number of photons presented in the core qubit states $c_0|0\rangle + c_1|1\rangle$ but it does not cause achieving of the following non-Gaussian attribute typical for Fock state [2].

The criteria will be derived ab initio without any assumptions about inspected states of light. Thus, they can be applied on any states with any mean number of photons. As such the criteria depend only on characteristics of detection process. The witnessing of condition (1) for an arbitrary state is provided by a completely characterized multichannel detector. Incoming light is evenly split to $n+1$ single-photon avalanche diodes (SPADs) that distinguish signal from vacuum. A genuine $n$ order property in (1) is detected when probability of simultaneous click of all $n+1$ SPADs (error) is suppressed sufficiently relative to the probability of simultaneous $n$ clicks (success). Let us choose any set of $n$ detectors and define the probability of their simultaneous detection by $P_s$ and the probability of all $n+1$ detectors clicking by $P_e$. In this case, $P_s$ refers to the probability of an expected success event, when light contains at least $n$ photons, and $P_e$ quantifies the probability of an unwanted error event, when light contains at least $n+1$ photons. Linear combination of both probabilities

$$F_{a,n}(\rho) = P_s + aP_e,$$

where $a$ is a free parameter, identifies the quantum attribute (1) if

$$\exists a : P_s + aP_e > F_{n}(a),$$

where $F_n(a)$ is a threshold function that is determined from optimizing functional $F_{a,n}(\rho)$ over mixtures of states given by the right side of (1). The subscript $n$ denotes the number of SPADs required for a success event. Because the functional is linear in a state, the optimum is obtained as a pure state $S(\beta)D(\alpha)|\tilde{\psi}_{n-1}\rangle$ where

$$|\tilde{\psi}_{n-1}\rangle = \sum_{k=0}^{n-1} c_k |k\rangle.$$  

The state is formally expressed by $2n+4$ parameters which hold normalization. Since two states with different global phases are identical, the considered state is determined by $2(n+1)$ unique parameters. The task is finding an optimum over these parameters. This can be performed only numerically. An algorithm which searches for the optimum has to incorporate extensive formulas that express general parametrization of success and error probabilities [30]. However, assuming that inspected states have strongly suppressed probability of error $P_e$, as is typical for high quality multiphoton states, the threshold can be derived at least approximately, see Supplementary Material.

The numerical optimizing could be done with several simplifying assumptions which were verified by Monte-Carlo simulation. The result is that the optimal state is parametrized by real parameters $\alpha$, $\beta$, $c_0$, ..., $c_{n-1}$. It
Figure 2: The faithful hierarchy witnessing the genuine n-photon quantum non-Gaussianity up to order three and its experimental verification. The quantum non-Gaussianity is recognized in the orange regions. The approximate solutions of the thresholds for n = 1, 2, 3 are plotted by dashed lines. The reliability of the thresholds is demonstrated by results of Monte-Carlo simulation. The gray points represent fifty points closest to the threshold that were generated in the simulation in a region close to cross section of measured data with the threshold after noise addition. The total number of runs in the simulation was 10⁶ (2nd order) and 10⁸ (3rd order). The black points correspond to the experimental data. The shifting of the points along the vertical axes corresponds to deterioration of the emitted light by background Poissonian noise. The slight movement of the points in the horizontal axis is caused by experimental imperfections resulting in noise leakage into the heralding arm. The mean number of photons of the background noise registered in a detection window is \(\bar{n}\) = 0, 4 \times 10⁻², 2 \times 10⁻⁴, 10⁻¹ (from the lower points to the upper points) for each measurement. Error bars represent statistical error of the number of detected coincidence events. The blue lines indicate theoretical estimate of attenuation on the experimental data. Corresponding values (the lower points to the upper points) for each measurement. Error bars represent statistical error of the number of detected coincidence events. The blue lines indicate theoretical estimate of attenuation on the experimental data. Corresponding values (the lower points to the upper points) for each measurement. Error bars represent statistical error of the number of detected coincidence events. The blue lines indicate theoretical estimate of attenuation on the experimental data. Corresponding values (the lower points to the upper points) for each measurement. Error bars represent statistical error of the number of detected coincidence events. The blue lines indicate theoretical estimate of attenuation on the experimental data. Corresponding values (the lower points to the upper points) for each measurement. Error bars represent statistical error of the number of detected coincidence events.

Let us first search for an optimal squeezing and displacement to maximize \(F_\alpha\) for a fixed core state \(|\tilde{\psi}_{n-1}\rangle\). The optimum satisfies \(\partial_\beta F_\alpha(\alpha, \beta, |\tilde{\psi}_{n-1}\rangle) = 0\) and \(\partial_\alpha F_\alpha(\alpha, \beta, |\tilde{\psi}_{n-1}\rangle) = 0\). Excluding the free parameter \(a\) from both equations leads to
\[
\partial_\alpha P_s \partial_\beta P_e - \partial_\beta P_s \partial_\alpha P_e = 0.
\]
It determines a function \(A(\beta, |\tilde{\psi}_{n-1}\rangle)\) that binds the displacement with the other parameters
\[
|\alpha|^2 := A(\beta, |\tilde{\psi}_{n-1}\rangle).
\]
Since the task employs searching for the maximal probability of success with constraint on the error probability, the squeezing can be always adjusted so that the probability of error gains the constraint value. This unambiguously determines the optimal squeezing and displacement of a core state \(|\tilde{\psi}_{n-1}\rangle\) so that the final state has the maximal probability of success. Then, it remains to optimize over the core states \(|\tilde{\psi}_{n-1}\rangle\) to obtain the threshold witnessing the desired quantum attributes. The derived thresholds are depicted in Fig. 2 for layouts with three and four detectors. The thresholds in the log-log scale can be approximated by linear functions satisfying
\[
P_e \approx \frac{(1 + n)^{2n}(2 + n)^2(1 + n)!P_s^3}{18n^2(n!)^3}.
\]
These approximations are applicable as a basic witness, however, they are below the real thresholds. They have to be carefully used if data are very close to them, surpassing them too tightly can lead to a false positive. Thus, the Supplementary Material provides derivation of more accurate approximations that can be applied on a larger set of states.

Let us note that although the thresholds were derived from an assumption of single mode states, they can be applied even on detection of states occupying more modes. The genuine n-photon quantum non-Gaussianity of multi-mode states means the higher photon contributions are produced neither by squeezing nor by displacement of multi-mode core state that shows truncated photon distribution. Since the exact definition of that property is technical in the multi-mode case, it is presented in Supplementary Materials. Also, the Supplementary Material describes details of Monte-Carlo simulation indicating the thresholds do not get stricter forms when they cover the refused multi-mode states instead of the single mode ones.

Experimental setup. — To experimentally witness genuine n-photon quantum non-Gaussianity, a superior multi-photon source is required. In this regard, there has been recently reported development in the works [31–33]. We generated statistics with controllable multi-photon content from a well-established photon source based on multiple high-quality single photons triggered to suppress random noise. We employed continuous-wave parametric down-conversion (SPDC) to generate sequences of n heralded single photons that were collectively measured on a multichannel detector (see Fig. 4). In addition to this signal, we added extra Poissonian background noise from a laser diode to explore the sensitivity of genuine n-photon quantum non-Gaussianity to multiphoton content. We detected all photons from all time modes collectively, as determined by triggering events and detector resolution, considering the overall statistics.
The source of heralded single photons consisted of a periodically poled KTP pumped by a 405-nm laser diode and set to a collinear type-II configuration. The detector was implemented by a balanced network of half-wave plates and polarizing beam splitters with a silicon single-photon avalanche diode (SPAD) in each arm. We recorded coincidence events between individual SPADs and obtained results presented in Figs. S2 and 3.

Results and analysis.— The data exhibit genuine quantum features up to order three \((n = 3)\). This was achieved by minimizing SPDC gain and the time window for coincidence detection, because \(P_e\) grows linearly with both parameters. The time window is limited by the temporal resolution of the detectors, while the gain can be lowered arbitrarily at the cost of reducing generation rate. The experimental limit of our demonstration was the measurement time needed to acquire statistically significant results for \(P_e\). The scaling is very fast: while we needed only 16 hours to obtain the results for \(n = 3\), several months would be needed for \(n = 4\). The main factor is that lowering SPDC gain simultaneously decreases the portion of error events and generation rate. A low event rate limits similarly also other experiments demonstrating negative Wigner function of a three-photon state \([10, 42, 43]\). To maintain a sufficiently low error rate with an increased gain, the detection time window would have to be reduced. This parameter is limited by the temporal resolution of SPADs and could be augmented by using detectors optimized for low jitter. Optical loss in both arms of the source, including detection loss, is also a factor, which depends on coupling efficiency as well as detector efficiency. The final limiting factor are the background dark counts, which become relevant in the extremal case of a very low gain and long measurement. Overall, the detection precision, signal-to-noise ratio and efficiency represent the main factors in the presented type of measurement. Fig. S2 shows that robustness against losses and noise rapidly decreases with higher order. This is a consequence of decreasing the maximum gain allowable for higher \(n\). Our data were all measured with the same gain, which means the individual statistics of all constituent heralded events are the same. The relation between number of heralded events and successful witnessing of genuine \(n\)-photon quantum non-Gaussianity is presented in Fig. 3. If the number of heralded events exceeds order of the witnessing criterion, the detection of
that property in our measurement fails. Note, this observation is not guaranteed by the definition \cite{1} of the hierarchy. When the order of a criterion is greater than a number of heralded events, the property is not detected from definition. These cases are however not depicted in the Fig. \ref{fig:3} due to the scale of confidence intervals of relevant error and success probabilities.

**Outlook.**—The presented hierarchy of quantum non-Gaussianity for the states approaching Fock states of light and its experimental verification can be applied to a class of new multiphoton experiments \cite{28} and to observe quantum non-Gaussianity of first photonic triplets \cite{29} from cubic nonlinear materials \cite{30–41}. As such, it can stimulate further experimental research in this pioneering direction of quantum technology with multiphoton states of light. Ab initio approach to hierarchy allows further extensions towards different quantum non-Gaussian states and its multi-mode versions used for both fundamental tests \cite{44,45} as well for applications in quantum technology with light \cite{46,47}. Because light is dominantly used for read out of atomic and solid state systems, this methodology can be used and also extended to evaluate quantum non-Gaussianity of, for example, already developed atomic-ensemble memories \cite{48–51} and new single-phonon mechanical oscillators \cite{52}.

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Supplementary information: Faithful hierarchy of
genuine \( n \)-photon quantum non-Gaussian light

The formula (1) in the main part of the manuscript
defines a hierarchy of quantum non-Gaussian properties
only for single mode states. The genuine \( n \)-photon
quantum non-Gaussianity recognize statistically signifi-
cant highly nonclassical features of light and thus it can
be extended even for multi-mode states. Let \( M \) denotes
number of considered modes. Similarly as in the single
mode case, the extended definition involves a statistically
truncated core state \( |\tilde{\psi}_{n-1}\rangle \) that exhibits

\[
\langle m_1 \rangle \otimes \ldots \otimes \langle m_M \rangle |\tilde{\psi}_{n-1}\rangle \neq 0 \quad (S1)
\]

only if \( \sum_{i=1}^{M} m_i < n \), where \( \langle m_i \rangle \) is a Fock state \( m \) oc-
ccupying the \( i \)th mode. The constrain guarantees the
state does not produce \( n \) or more than \( n \) photons. In
a single mode case, the core state can be expressed as
\( |\tilde{\psi}_{n-1}\rangle = \sum_{k=0}^{n-1} c_k |k\rangle \). Two modes core states correspond to
\( |\tilde{\psi}_{n-1}\rangle = \sum_{k=0}^{n-1} \sum_{l=0}^{n-k-1} C_{k,l} |k,l\rangle \) with arbitrary co-
efficients \( C_{k,l} \). The higher photon contributions of the
refused states can be generated only by squeezing or dis-
placement influencing all modes occupied by \( |\tilde{\psi}_{n-1}\rangle \). Let
\( S_i(\beta_i) \) be squeezing and \( D_i(\alpha_i) \) be displacement affecting
mode \( i \), where \( \beta_i \) and \( \alpha_i \) are parameters determining the
operators. A pure multi-mode state \( |\psi\rangle \) exhibits genuine
\( n \)-photon quantum non-Gaussianity if

\[
|\psi\rangle \neq S_M(\beta)D_M(\alpha)|\tilde{\psi}_{n-1}\rangle , \quad (S2)
\]

where \( \alpha \) and \( \beta \) are vectors \( \alpha = (\alpha_1, \ldots, \alpha_M) \), \( \beta =
(\beta_1, \ldots, \beta_M) \) and \( S_M(\beta) \), \( D_M(\alpha) \) are defined as tensor
products

\[
S_M(\beta) = \Pi_{i=1}^{M} S_i(\beta_i) ,
D_M(\alpha) = \Pi_{i=1}^{M} D_i(\alpha_i) . \quad (S3)
\]

The genuine \( n \)-photon quantum non-Gaussianity also ref-
uses all statistical mixtures of right side of inequality \( S2 \). It remains to check that function \( F_n(\alpha) \) covers all
states parametrized by the right side of inequality \( S2 \).
A partial proof can be find for states with low probabil-
ity of error. The only general approach is Monte-Carlo
simulation which, however, requires further software de-
velopment to be performed due to a large number of
parameters determining a general multi-mode \( n \)-photon
Gaussian state. The simulation for two mode states was
carried out only with real coefficient of the core state
\( |\tilde{\psi}_{n-1}\rangle \) and squeezing orthogonal to displacement. That
decreases a number of parameters over which the optimi-
tum is searched to \( \frac{1}{2}n(n+1) + 1 \). The results are pre-
sented in Fig. S1 for the second and third order of the
criteria.

The limit of states with low probability of error in-
volves states with small squeezing and small displace-
ment. Then, the error and success probabilities can be

\[
A(V, |n - 1\rangle) \approx 8\beta + \frac{16}{3} (3 + 2n + n^2 )\beta^2 \quad (S6)
\]

where \( 0 < \beta \ll 1 \). Inserting it into formulas \( S4 \) results in approximates

\[
P_e \approx \frac{mn!(n+2)^2}{55296(n+1)^{n+1}} \beta^3 \left[ 384 + \beta(896 + 307n + 99n^2) \right] ,
P_s \approx \frac{mn!}{12(1+n)^n} \beta \left[ 6 + (6 + 2n + n^2 )\beta \right] . \quad (S7)
\]
where $t$ substitutes the squeezing by $t = 1 - V$. Those expressions parametrize the thresholds tightly even beyond the considered regime of weak states and they can be used as a better approximate. Their reliability was checked by Monte Carlo simulation up to order five. The results of simulation related to the fourth and fifth ordered criterion are illustrated in Fig. S2. Assuming $t \ll 1$, the criteria get simpler forms

$$P_e < \frac{(1 + n)^2 (2 + n)^2 (1 + n)! P_s^3}{18 n^2 (n!)^3},$$  \hspace{1cm} \text{(S8)}$$

which however diverge quickly from the exact threshold when $P_e$ grows.

The experimental demonstration of genuine $n$-photon quantum non-Gaussianity is completed by information about robustness against losses and additional noise, which can be theoretically predicted in the region of states with low probability of error. In those cases, a click statistics is contributed approximately only by

$$P_s \approx \frac{n!}{(n + 1)^n} \rho_n$$
$$P_e \approx \frac{n!}{(n + 1)^n} \rho_{n+1},$$  \hspace{1cm} \text{(S9)}$$

where $\rho_k$ is a probability a state $\rho$ has $k$ photons, i.e. $\rho_k = \langle k | \rho | k \rangle$. Transmission efficiency $T$ changes the photon statistics approximately so this $\rho_k \rightarrow \rho_k T^k$. In the

\[ \log_{10} P_e = \log_{10} P_e^{(T)} = \frac{n + 1}{n} \left( \log_{10} P_s^{(T)} - \log_{10} P_s + \log_{10} P_e \right), \]  \hspace{1cm} \text{(S10)}$$

where $P_s^{(T)}$ are measured probabilities affected by attenuation and $P_s, P_e$ are the original ones. Employing this rule together with (S8) enable to estimate the robustness against losses. Poissonian noise influence the statistics by

$$P_s \approx \frac{n!}{(n + 1)^n} (\rho_n + \bar{n} \rho_{n-1})$$
$$P_e \approx \frac{n!}{(n + 1)^n} (\rho_{n+1} + \bar{n} \rho_n),$$  \hspace{1cm} \text{(S11)}$$

where $\bar{n}$ is a mean number of photons of the noise. Because the criteria impose a very strict condition on the noise, one can assume the members contributing the success probability satisfy $\bar{n} \rho_{n-1} \ll \rho_n$. Therefore a point moves in the plots vertically along the $\log_{10} P_e$ axis. However, the data show slight dropping of probability $P_s$ for states affected by the background noise, as apparent in the Fig. 2 of the main part of the manuscript. It arises from imperfect protection of the heralding detector from photons of the noise in the experiment. Thus, a heralding event was rarely caused by the background noise that decrease slightly probability $P_s$.

[1] J. Sperling, W. Vogel, and G. S. Agarwal, Phys. Rev. A 85, 023820 (2012)