Nonclassicality and entanglement criteria for bipartite optical fields characterized by quadratic detectors

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Numerous inequalities involving moments of integrated intensities and revealing nonclassicality and entanglement in bipartite optical fields are derived using the majorization theory, non-negative polynomials, the matrix approach, as well as the Cauchy-Schwarz inequality. Different approaches for deriving these inequalities are compared. Using the experimental photocount histogram generated by a weak noisy twin beam monitored by a photon-number-resolving iCCD camera the performance of the derived inequalities is compared. A basic set of ten inequalities suitable for monitoring the entanglement of a twin beam is suggested. Inequalities involving moments of photocounts (photon numbers) as well as those containing directly the elements of photocount (photon-number) distributions are also discussed as a tool for revealing nonclassicality.

I. INTRODUCTION

The notion of a nonclassical field has been rigorously defined once the famous Glauber-Sudarshan representation of the density matrix of an optical field was formulated [1, 2]. From that time, any optical field with a non-positive Glauber-Sudarshan quasi-distribution is considered as nonclassical [3–6]. The analysis of more complex optical fields involving several optical modes has shown that one of the reasons for field’s nonclassicality is the presence of quantum correlations (entanglement) among the modes that constitute the field. As the entanglement is interesting both for fundamental reasons and various applications (in metrology, quantum-key distribution, etc.), it has been extensively studied in the last tens’ years in numerous publications. The simplest case of entanglement between two fields has naturally attracted the greatest attention. In this case, even the quantification of entanglement has been found using the Schmidt number for pure states [7] and its generalization to mixed states based on finding the closest pure entangled state. Also an alternative quantification derived from the shape of the Wigner function has been given [8]. Unfortunately, these theoretical approaches are difficult to be applied to experimental optical fields [9, 10]. From the experimental point of view, joint homodyne tomography [11, 12] of both fields is needed to reveal the joint phase-space quasi-distribution of these fields and, subsequently, quantify the entanglement via the mentioned theoretical approaches. Large experimental demands of entanglement quantification lead to a simpler concept of entanglement witnesses (criteria) when dealing with the entanglement. An entanglement witness is a physical quantity which identifies the entanglement qualitatively through its values. Typically, such quantity is constructed from an inequality fulfilled by any classical optical field. A well-known and frequently-used PPT criterion [13, 14] represents an entanglement witness that exploits the eigenvalues of a certain matrix. For specific systems, this witness can even be converted into an entanglement measure called negativity [15]. There exists in principle an infinite number of entanglement witnesses. On the other hand, some of these witnesses are more important (or useful) for the physical reasons. These reasons are pragmatic and they are related to their performance in the experimental characterization of optical fields. As quadratic optical detectors are by-far the most frequently used detectors in optical laboratories worldwide, the witnesses exploiting the moments of integrated intensity (farther only intensity) are extraordinarily important [16–20]. We note that the measurement of the whole joint photocount distribution of a bipartite optical field can be used to reconstruct the joint quasi-distribution of integrated intensities [21] and to reveal its negative values observed for nonclassical states.

Here, we theoretically as well as experimentally analyze the witnesses that indicate negative values of the Glauber-Sudarshan quasi-distribution of intensities. When applied to the whole optical field they represent global nonclassicality criteria (GNCCa). On the other hand, they serve as local nonclassicality criteria (LNCCa) in the cases of marginal fields describing individual optical modes. For bipartite optical fields with classical constituents, the GNCCa represent also entanglement between the constituents or both. Twin beams with their signal and idler

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beams containing many photon pairs represent a typical example of such bipartite optical fields. The GNCCa and LNCCa are derived by several approaches that use the majorization theory \(^{22}\), consider non-negative polynomials and quadratic forms (the matrix approach) \(^{23, 24}\) and exploit the Cauchy-Schwarz inequality. Relying on the Mandel photodetection formula \(^{25}\), the corresponding inequalities among the elements of the joint photocount and photon-number distributions are also revealed. The performance of the derived GNCCa is tested on the experimental data characterizing a twin beam with around nine photon pairs on average and acquired by an intensified CCD (iCCD) camera. In this case, the GNCCa are also entanglement criteria.

The paper is organized as follows. In Sec. II, we give the simplest inequalities among intensity moments. More complex inequalities including multiple intensity moments are derived in Sec. III using different approaches. Inequalities using the elements of the joint photocount and photon-number distributions are discussed in Sec. IV, together with some useful inequalities containing photocount and photon-number moments. Sec. V is devoted to the application of the derived inequalities to an experimental noisy twin beam. Conclusions are drawn in Sec. VI. Additional inequalities for identifying nonclassicality, that are redundant to those written in the main text, are summarized in Appendix A for completeness.

II. SIMPLE NONCLASSICALITY CRITERIA USING INTENSITY MOMENTS

We consider a bipartite optical field composed of in general two entangled fields, that we call the signal and idler fields and that have intensities \(W_s\) and \(W_i\), respectively. The overall field is described by the joint signal-idler intensity quasi-distribution \(P_{si}(W_s, W_i)\) \(^{22}\) that allows to determine the normally-ordered (intensity) moments \(^{3}\) along the relation:

\[
\langle W_s^k W_i^l \rangle = \int_0^\infty dW_s \int_0^\infty dW_i W_s^k W_i^l P_{si}(W_s, W_i),
\]

\[k, l = 0, 1, \ldots \]

(1)

According to the majorization theory applied to polynomials written in two independent variables \(^{22, 26}\), these intensity moments fulfill certain classical inequalities. Their negation gives us the following series of Global NonClassicality Criteria:

\[
\sum_{\{k,l\}} \langle W_s^k W_i^l \rangle < \sum_{\{k',l'\}} \langle W_s^{k'} W_i^{l'} \rangle
\]

(2)

where the summation is performed over all possible permutations of the indices and the indices \(k,l\) majorize the indices \(k',l'\) (\(\{k,l\} > \{k',l'\}\)). We note that such GNCCa are obtained in a general form of sum (and difference) of mean values.

To understand in detail the structure of such GNCCa, we explicitly write those containing the intensity moments up to the fifth order in the form that naturally arises in the majorization theory:

\[
\langle W_s^2 \rangle + \langle W_i^2 \rangle < 2\langle W_s W_i \rangle,
\]

(3)

\[
\langle W_s^3 \rangle + \langle W_i^3 \rangle < \langle W_s^2 W_i \rangle + \langle W_s W_i^2 \rangle,
\]

(4)

\[
\langle W_s^4 \rangle + \langle W_i^4 \rangle < \langle W_s^2 W_i^2 \rangle + \langle W_s W_i^3 \rangle + \langle W_i W_s^3 \rangle,
\]

(5)

\[
\langle W_s^4 \rangle + \langle W_i^4 \rangle < 2\langle W_s^2 W_i^2 \rangle,
\]

(6)

\[
\langle W_s^5 \rangle + \langle W_i^5 \rangle < 2\langle W_s^2 W_i^3 \rangle,
\]

(7)

\[
\langle W_s^5 \rangle + \langle W_i^5 \rangle < \langle W_s^2 W_i^3 \rangle + \langle W_s W_i^4 \rangle + \langle W_i W_s^4 \rangle,
\]

(8)

\[
\langle W_s^5 \rangle < \langle W_s^2 W_i^3 \rangle + \langle W_s W_i^4 \rangle + \langle W_i W_s^4 \rangle,
\]

(9)

\[
\langle W_s^4 W_i \rangle + \langle W_s W_i^4 \rangle < \langle W_s^2 W_i^2 \rangle + \langle W_s^2 W_i^2 \rangle.
\]

(10)

However, the inequalities in Eqs. (3) – (10) can be recast in turn into the following ones:

\[
\langle W_s - W_i \rangle^2 < 0,
\]

(11)

\[
\langle W_s + W_i \rangle \langle W_s - W_i \rangle \langle W_s^2 - W_i^2 \rangle < 0,
\]

(12)

\[
\langle W_s^2 + W_s W_i + W_i^2 \rangle \langle W_s - W_i \rangle^2 < 0,
\]

(13)

\[
\langle W_s^2 + 2W_s W_i + W_i^2 \rangle \langle W_s^2 - W_i^2 \rangle < 0,
\]

(14)

\[
\langle W_s W_i \rangle \langle W_s - W_i \rangle^2 < 0,
\]

(15)

\[
\langle W_s + W_i \rangle \langle W_s^2 + W_s W_i + W_i^2 \rangle \langle W_s - W_i \rangle^2 < 0,
\]

(16)

\[
\langle W_s + W_i \rangle \langle W_s^2 + W_s W_i + W_i^2 \rangle \langle W_s - W_i \rangle^2 < 0.
\]

(17)

A common property of these inequalities is that they are symmetric with respect to the exchange of indices \(s\) and \(i\). This has its origin in the majorization theory.

Natural generalization of the above GNCCa that removes this symmetry and that is based upon mean values of non-negative polynomials is written in the form of the following Global NonClassicality Criteria:

\[
\langle W_s^k W_i^l (W_s - W_i)^{2m} \rangle < 0,
\]

\[k, l, m = 0, 1, \ldots, m = 1, 2, \ldots \]

(19)

Considering \(m = 1\) in Eq. (19) and intensity moments up to the fifth order, we may define the following GNCCa \(E\):

\[
E_{001} \equiv \langle W_s^2 \rangle + \langle W_i^2 \rangle - 2\langle W_s W_i \rangle < 0,
\]

(20)

\[
E_{011} \equiv \langle W_s^3 \rangle + \langle W_s W_i^2 \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(21)

\[
E_{011} \equiv \langle W_s^3 \rangle + \langle W_i^2 W_s \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(22)

\[
E_{201} \equiv \langle W_s^3 \rangle + \langle W_s^3 W_i^2 \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(23)

\[
E_{201} \equiv \langle W_i^3 \rangle + \langle W_s^2 W_i^2 \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(24)

\[
E_{111} \equiv \langle W_s^3 W_i \rangle + \langle W_s W_i^3 \rangle - 2\langle W_s^2 W_i^2 \rangle < 0,
\]

(25)

\[
E_{301} \equiv \langle W_s^3 \rangle + \langle W_s^3 W_i^2 \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(26)

\[
E_{301} \equiv \langle W_s^3 \rangle + \langle W_s^2 W_i^3 \rangle - 2\langle W_s^2 W_i \rangle < 0,
\]

(27)

\[
E_{211} \equiv \langle W_s^4 W_i \rangle + \langle W_s^3 W_i^2 \rangle - 2\langle W_s^2 W_i^2 \rangle < 0,
\]

(28)

\[
E_{211} \equiv \langle W_s^4 W_i \rangle + \langle W_s^3 W_i^2 \rangle - 2\langle W_s^2 W_i^2 \rangle < 0.
\]

(29)

The original GNCCa given in Eqs. (3) — (10) represent a subset of the GNCCa written in Eqs. (20) — (29). In detail, the GNCCa in Eqs. (3) — (10) are expressed in
turn as $E_{001}$, $E_{101}$, $E_{201}$, $E_{011}$, $E_{211}$, $E_{111}$, $E_{202}$, $E_{021}$, $E_{212}$, $E_{121}$, $E_{203}$, $E_{031}$, $E_{213}$, $E_{121}$, $E_{204}$, $E_{042}$, $E_{221}$, $E_{212}$, $E_{122}$ and $E_{205}$.

Moreover, the consideration of $m = 2$ in Eq. (19) gives us additional three GNCCa:

\[
E_{002} \equiv \langle W_s^2 \rangle - 4\langle W_s^3 W_i \rangle + 6\langle W_s^2 W_i^2 \rangle - 4\langle W_s W_i^3 \rangle + \langle W_i^4 \rangle < 0,
\]

\[
E_{102} \equiv \langle W_s^2 \rangle - 4\langle W_s W_i \rangle + 6\langle W_s^2 W_i^2 \rangle - 4\langle W_s^2 W_i^3 \rangle + \langle W_i^4 \rangle < 0,
\]

\[
E_{012} \equiv \langle W_s^3 W_i \rangle - 4\langle W_s^3 W_i^2 \rangle + 6\langle W_s^2 W_i^3 \rangle - 4\langle W_s W_i^4 \rangle + \langle W_i^5 \rangle < 0.
\]

These GNCCa can be expressed as linear combinations of some of the GNCCa written in Eqs. (20)–(29):

\[
E_{002} = E_{201} + E_{021} - 2E_{111},
\]

\[
E_{102} = E_{301} + E_{121} - 2E_{211},
\]

\[
E_{012} = E_{211} + E_{031} - 2E_{121}.
\]

As negative signs occur in the combinations of GNCCa $E$ on the right-hand-sides of Eqs. (33), the GNCCa $E_{002}$, $E_{102}$ and $E_{012}$ are nontrivial and enrich the set of GNCCa given in Eqs. (20)–(29). We note that analogical situation is met for $m > 2$ in Eq. (19) and higher-order intensity moments.

III. NONCLASSICALITY CRITERIA CONTAINING MULTIPLE INTENSITY MOMENTS

In this section, we derive the non-classicality criteria that involve products of intensity moments. We concentrate our attention to the GNCCa containing products of two intensity moments, though several GNCCa encompassing also products of three intensity moments are mentioned. To determine such GNCCa we first apply the majorization theory. Then we exploit non-negative polynomials to arrive at additional GNCCa. For completeness, we mention the GNCCa reached by the matrix approach, that uses non-negative quadratic forms, and those derived from the Cauchy-Schwarz inequality. In parallel, we also reveal LNCCa containing intensity moments and provided by the majorization theory.

A. Nonclassicality criteria based on the majorization theory

We use again the formulas of the majorization theory \cite{22}, now in the systematic way. We begin with the majorization theory applied to polynomials written in two independent variables $W_s$ and $W_i$. Contrary to the approach of the previous section, we make averaging with the factorized quasi-distribution function $P_s(W_s)P_i(W_i)$ where $P_s$ ($P_i$) stands for the signal (idler) reduced quasi-distribution function. The original Eq. (2) attains in this case the form of the following Local NonClassicality Criteria:

\[
\sum_{(k,l)} \langle W_s^k \rangle \langle W_i^l \rangle < \sum_{(k',l')} \langle W_s^{k'} \rangle \langle W_i^{l'} \rangle; \quad (34)
\]

where $\{k, l\} \neq \{k', l'\}$. Considering intensity moments up to the fifth order, we arrive at the following six LNCCa expressed in terms of the intensity moments of the local signal and idler fields:

\[
B_{11}^{20} \equiv \langle W_s^2 \rangle + \langle W_s^2 \rangle - 2\langle W_s \rangle \langle W_i \rangle < 0,
\]

\[
B_{21}^{20} \equiv \langle W_s^2 \rangle + \langle W_s^3 \rangle - \langle W_s^2 \rangle \langle W_i \rangle - \langle W_s \rangle \langle W_i^2 \rangle < 0, \quad (35)
\]

\[
B_{31}^{20} \equiv \langle W_s^3 \rangle + \langle W_s^3 \rangle - \langle W_s^2 \rangle \langle W_i \rangle - \langle W_s \rangle \langle W_i^2 \rangle < 0, \quad (36)
\]

\[
B_{41}^{20} \equiv \langle W_s^4 \rangle + \langle W_s^3 \rangle - \langle W_s^3 \rangle \langle W_i \rangle - \langle W_s \rangle \langle W_i^3 \rangle < 0, \quad (37)
\]

\[
B_{51}^{20} \equiv \langle W_s^5 \rangle + \langle W_s^4 \rangle - \langle W_s^4 \rangle \langle W_i \rangle - \langle W_s \rangle \langle W_i^4 \rangle < 0, \quad (38)
\]

\[
B_{61}^{20} \equiv \langle W_s^6 \rangle + \langle W_s^5 \rangle - \langle W_s^5 \rangle \langle W_i \rangle - \langle W_s \rangle \langle W_i^5 \rangle < 0. \quad (39)
\]

We note that the LNCCa given in Eqs. (35)–(39) occur in more complex expressions derived below, that combine the local nonclassicalities with the entanglement. We also note that the simplest LNCC given in Eq. (41) was experimentally observed already in 1977 using the light from fluorescence of a single molecule \cite{31}.

To reveal more complex GNCCa, we first analyze the formulas of the majorization theory with three independent variables $W_s$, $W_i$ and $W_a$ considering two kinds of averaging with the quasi-distribution functions $P_a(W_s, W_i)P_a(W_a)$, $a = i, s$. To demonstrate the structure of the obtained inequalities without treating more complex formulas, we investigate the inequalities including intensity moments up to the fourth order. Detailed analysis of the majorization formulas denoted in the standard notation as $\{200\} \Rightarrow \{110\}, \{300\} \Rightarrow \{210\}, \{400\} \Rightarrow \{310\}$, and $\{310\} \Rightarrow \{220\}$ reveals that all these inequalities are obtained as suitable positive linear combinations of some of the inequalities written in Eqs. (20)–(29) and \cite{35}–\cite{40} and so they are redundant for the indication of nonclassicality. They can be found in Appendix A [Eqs. (A17)–(A20)]. The remaining majorization inequalities $\{210\} \Rightarrow \{111\}$ and $\{220\} \Rightarrow \{211\}$ considered
with both types of averaging then provide the following four Global NonClassicality Criteria \((a = s, i)\):

\[
a D_{111}^{10} = 2\langle W_a^s \rangle \langle W_a \rangle + \langle W_s^2 W_i \rangle + \langle W_s^2 W_i^2 \rangle + \langle W_s \rangle \langle W_i^2 \rangle + \langle W_s \rangle \langle W_i^2 \rangle - 6\langle W_a \rangle \langle W_a \rangle < 0, \tag{47}
\]

\[
a D_{211}^{220} = (\langle W_s^2 \rangle^2 + \langle W_s^2 W_i \rangle + \langle W_s^2 \rangle \langle W_i^2 \rangle - \langle W_a \rangle \times [\langle W_s^2 \rangle + \langle W_s \rangle \langle W_i \rangle]^2 - 6\langle W_a^s \rangle \langle W_a \rangle < 0. \tag{48}
\]

In the next step, we analyze the majorization inequalities with four independent variables \(W_s, W_i, W_s', \text{ and } W_i'\) and we use the quasi-distribution function \(P_a(W_s, W_i) P_a(W_s', W_i')\) for averaging. The inequalities \([2000] \succ [1110], [3000] \succ [2100], [4000] \succ [3100],\) and \([3100] \succ [2200]\) can be expressed as positive linear combinations of those given in Eqs. \([20] - [29]\) and \([35] - [46]\) and as such they are not interesting for revealing nonclassicality. Similarly, the doubled inequality \([2100] \succ [1110], [2200] \succ [2110]\) is obtained as the sum \(a D_{111}^{210} + i D_{111}^{120} + i D_{220}^{220} + i D_{221}^{221}\) of the GNCCa written in Eq. \([47] - [48]\). More details are given in Appendix A [see Eqs. \([A2]-[A20]\)]. Only the inequality \([2110] \succ [1111]\) is recast into the following Global NonClassicality Criterion:

\[
D_{1111}^{1110} = [\langle W_s^2 \rangle \langle W_s \rangle + \langle W_s^2 W_i \rangle + \langle W_s \rangle \langle W_i \rangle + \langle W_s \rangle W_s'] \times [\langle W_s^2 \rangle + \langle W_s \rangle \langle W_i \rangle] - 6\langle W_a^s \rangle \langle W_a \rangle < 0. \tag{49}
\]

The remaining inequalities up to the fourth order are provided by the majorization inequalities \([2100] \succ [1110], [2200] \succ [2110]\) and \([2110] \succ [1111]\) if we perform in turn averaging with the following three quasi-distribution functions \(P_a(W_s, W_i) P_a(W_s', W_i') P_a(W_s', W_i')\), \(a = s, i\), and \(P_a(W_s, W_i) P_a(W_s', W_i') P_a(W_s', W_i')\). The occurrence of three intensity moments in a product represents their common feature. Step by step, the corresponding Global NonClassicality Criteria are derived in the form \((a = s, i)\):

\[
a T_{1111}^{210} = 6\langle W_s^2 \rangle \langle W_s \rangle + \langle W_s^2 W_i \rangle + \langle W_s^2 W_i^2 \rangle + 2\langle W_s \rangle \langle W_i \rangle + 2\langle W_s \rangle \langle W_i \rangle - 6\langle W_s \rangle \langle W_s \rangle < 0, \tag{50}
\]

\[
a T_{2110}^{220} = 2\langle W_s^2 \rangle \langle W_s \rangle + 2\langle W_s W_i \rangle + 2\langle W_s \rangle \langle W_i \rangle + 3\langle W_s \rangle \langle W_i \rangle + 3\langle W_s \rangle \langle W_i \rangle - 3\langle W_s \rangle \langle W_i \rangle \times \langle W_s^2 \rangle - 3\langle W_s \rangle \langle W_i \rangle < 0, \tag{51}
\]

\[
a T_{2210}^{220} = 6\langle W_s^2 \rangle^2 + 2\langle W_s^2 \rangle \langle W_s \rangle + 4\langle W_s \rangle \langle W_i \rangle - 2\langle W_s \rangle^2 \langle W_s \rangle - \langle W_s \rangle \langle W_i \rangle \langle W_s \rangle \langle W_i \rangle \times \langle W_s \rangle^2 \langle W_s \rangle - \langle W_s \rangle \langle W_i \rangle \langle W_s \rangle \langle W_i \rangle < 0, \tag{52}
\]

\[
a T_{2210}^{220} = 2\langle W_s^2 \rangle^2 + 2\langle W_s^2 \rangle^2 + 2\langle W_s^2 \rangle^2 + 6\langle W_s \rangle \langle W_i \rangle - 2\langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle 
- \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle - \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle < 0, \tag{53}
\]

\[
a T_{1111}^{2110} = 2\langle W_s \rangle \langle W_s \rangle + 2\langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle + \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle + \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \times \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle \langle W_s \rangle < 0, \tag{54}
\]

We note that the approach leading to Eqs. \([50] - [55]\) provides also additional redundant GNCCa that are summarized in Appendix A [see Eqs. \([A27]-[A34]\)].

Additional nonclassicality inequalities containing products of three intensity moments are reached from the majorization inequalities written for polynomials with three variables and assuming averaging with the factorized quasi-distributions \(P_s(W_s) P_s(W_i) P_s(W_s')\), \(a = s, i\). The majorization inequalities \([210] \succ [111]\) and \([220] \succ [211]\) leave us with the following Local NonClassicality Criteria in this case \((a = s, i)\):

\[
a B_{111}^{210} = \langle W_s^2 \rangle \langle W_s \rangle + \langle W_s^2 \rangle \langle W_i \rangle + \langle W_s^2 \rangle \langle W_s \rangle - 3\langle W_s \rangle \langle W_s \rangle < 0, \tag{56}
\]

\[
a B_{211}^{220} = \langle W_s^2 \rangle^2 + 2\langle W_s \rangle \langle W_s \rangle + \langle W_s \rangle^2 \langle W_s \rangle - \langle W_s \rangle^2 \langle W_s \rangle < 0. \tag{57}
\]

Analyzing the inequalities originating in the majorization theory with intensity moments up to the fourth order, we finally arrive at those written among the terms with four intensity moments in the product. They are naturally derived from the majorization inequalities written for polynomials with four variables considering in turn the quasi-distributions \(P_s(W_s) P_s(W_i) P_s(W_s') P_s(W_s')\), \(a = s, i\), and \(P_s(W_s) P_s(W_i) P_s(W_s') P_s(W_s')\). In detail, the majorization inequality \([2110] \succ [1111]\) is recast considering the above averaging into the following Local NonClassicality Criteria \((a = s, i)\):

\[
a B_{111}^{2110} = \langle W_s^2 \rangle \langle W_s \rangle + \langle W_s^2 \rangle \langle W_i \rangle + \langle W_s^2 \rangle \langle W_i \rangle - 4\langle W_s \rangle \langle W_s \rangle < 0, \tag{58}
\]

\[
a B_{111}^{2110} = \langle W_s^2 \rangle \langle W_i \rangle^2 + \langle W_s \rangle^2 \langle W_i \rangle^2 + 2\langle W_s \rangle + \langle W_i \rangle < 0. \tag{59}
\]

We note that also additional LNCa arise from the majorization theory written for polynomials with three and four variables. However, they can be expressed as positive linear combinations of the above written LNCa and so they are redundant. They are explicitly given in Eqs. \([A1]-[A16]\) in Appendix A.

**B. Nonclassicality criteria based on non-negative polynomials**

Similarly as in the previous section where we have used the mean values of non-negative polynomials in Eq. \([19]\), here we derive Local and Global NonClassicality Criteria
by negating the following classical inequalities:
\[ (W_s^k W_i^l (W_s - \langle W_s \rangle)^{2m} (W_i - \langle W_i \rangle)^{2n}) < 0, \]
\[ k, l = 0, 1, \ldots, m = n = 0, 1, \ldots. \]  
(60)

Concentrating on the signal field \((m = 1 \text{ and } n = 0)\) and restricting our attention to the LNCCa containing intensity moments up to the fifth order we recognize in Eqs. (60) the following LNCCa:
\[ E_{0010} \equiv \langle W_s^2 W_i^3 \rangle + \langle W_s^3 W_i^2 \rangle - 2\langle W_s \rangle \langle W_s W_i \rangle < 0, \]
\[ l = 1, 2, 3, \]  
(61)
\[ E_{1110} \equiv \langle W_s^2 W_i^4 \rangle + \langle W_s W_i^2 \rangle - 2\langle W_s \rangle \langle W_s W_i^2 \rangle < 0, \]
\[ l = 1, 2, \]  
(62)
\[ E_{2110} \equiv \langle W_s^3 W_i^2 \rangle + \langle W_s^2 W_i^3 \rangle - 2\langle W_s^2 \rangle \langle W_s W_i^3 \rangle < 0. \]  
(63)

One additional LNCC \((E_{0120})\) as well as one additional GNCC \((E_{0101})\) are expressed as linear combinations of the LNCCa in Eqs. (61)–(63) with varying signs:
\[ E_{0120} \equiv E_{2110} + \langle W_i^2 \rangle^2 E_{0110} - 2\langle W_s \rangle E_{1110} < 0, \]  
(64)
\[ E_{1011} \equiv E_{1210} + \langle W_i^2 \rangle^2 E_{1010} - 2\langle W_s \rangle E_{1110} < 0. \]  
(65)

The LNCCa and GNCC given in Eqs. (61)–(65) with exchanged subscripts \(s\) and \(i\) provide additional LNCCa and GNCC that can be derived from the symmetry. Moreover, there exists another GNCC belonging to the fourth order and being symmetric with respect to subscripts \(s\) and \(i:\)
\[ E_{0011} \equiv E_{0210} + \langle W_i^2 \rangle^2 \langle W_s^3 \rangle - 2\langle W_s \rangle E_{0110} < 0. \]  
(66)

We note that Eq. (60) considered for \(l = n = 0\) gives also nontrivial LNCCa that can be added to those written in Eqs. (61)–(66). They are expressed as:
\[ E_{1010} \equiv \langle W_i^2 \rangle \langle W_s \rangle^2 - \langle W_s \rangle^3 \langle W_i \rangle < 0, \]  
(67)
\[ E_{2010} \equiv \langle W_i^4 \rangle - \langle W_s \rangle \langle W_s \rangle^2 < 0, \]  
(68)
\[ E_{3010} \equiv \langle W_i^6 \rangle - \langle W_s \rangle \langle W_s \rangle^3 < 0, \]  
(69)
\[ E_{0202} \equiv \langle W_i^4 \rangle - 3\langle W_i^2 \rangle^2 \langle W_s \rangle^2 + 3\langle W_i \rangle^2 \langle W_s \rangle^2 \langle W_s \rangle < 0, \]  
(70)
\[ E_{1202} \equiv \langle W_i^6 \rangle - 3\langle W_s \rangle \langle W_i \rangle \langle W_s \rangle^2 + 3\langle W_s \rangle^2 \langle W_s \rangle < 0. \]  
(71)

C. Global nonclassicality criteria based on the matrix approach

In this case, the GNCCa are based on considering classically positive semi-definite matrices of dimension \(n \times n\) for \(n = 2, 3, \ldots\) that describe mean values of quadratic forms defined above the basis that includes different powers of the signal and idler intensities. This approach has been elaborated in general both for the amplitude and intensity moments in Refs. [23, 32, 54] summarized in Ref. 24 and applied in Ref. 19. The Bochner theorem has been used to arrive at the even more general forms of these inequalities [33, 36]. For \(n = 2\) the Global Nonclassicality Criteria are defined along the relation \((i, j, k, l \geq 0)\):
\[ M_{ijkl} \equiv \langle W_s^{2i} W_i^{2j} \rangle \langle W_s^{2k} W_i^{2l} \rangle - \langle W_s^{i+k} W_i^{j+l} \rangle^2 < 0. \]  
(72)

Restricting our considerations to the GNCCa up to the fifth order in intensity moments, we only reveal the following two inequalities:
\[ M_{1100} \equiv \langle W_s^2 W_i^2 \rangle - \langle W_s W_i \rangle^2 < 0, \]  
(73)
\[ M_{0101} \equiv \langle W_s^2 \rangle^2 - \langle W_s W_i \rangle^2 < 0. \]  
(74)

For comparison, we write down two GNCCa originat

D. Global nonclassicality criteria derived from the Cauchy-Schwarz inequality

To reveal additional Global Nonclassicality Criteria, we negate the Cauchy-Schwarz inequality:
\[ \left[ \int dW_i dW_i P_{si}(W_s, W_i) f(W_s, W_i) g(W_s, W_i) \right]^2 \]
\[ > \int dW_i dW_i P_{si}(W_s, W_i) f^2(W_s, W_i) \]
\[ \times \int dW_s dW_i P_{si}(W_s, W_i) g^2(W_s, W_i). \]  
(78)

In Eq. (78), \(f\) and \(g\) denote arbitrary real functions and \(P_{si}\) stands for the joint quasi-distribution of integrated intensities. Restricting ourselves up to the fifth power of intensities, we may in turn consider \(f = 1\) together with \(g = W_s W_i, f = \sqrt{W_s} W_i\) together with \(g = \sqrt{W_s} W_i, f = W_s\) together with \(g = W_i\), and \(f = W_s \sqrt{W_i}\) together with \(g = \sqrt{W_i} \) to arrive at the following GNCCa:
\[ C_{00}^{00} \equiv \langle W_s^2 W_i^2 \rangle - \langle W_s W_i \rangle^2 < 0, \]  
(79)
\[ C_{10}^{00} \equiv \langle W_s^3 W_i^2 \rangle - \langle W_s W_i \rangle^2 < 0, \]  
(80)
\[ C_{00}^{20} \equiv \langle W_s^2 \rangle^2 - \langle W_s W_i \rangle^2 < 0, \]  
(81)
\[ C_{00}^{30} \equiv \langle W_s^4 \rangle - \langle W_s W_i \rangle^2 < 0. \]  
(82)

The criterion \(C_{22}^{00}\) in Eq. (79) \(C_{20}^{00}\) in Eq. (81) coincides with the criterion \(M_{1100}\) in Eq. (73). \(M_{0001}\) in Eq. (74) derived from the matrix approach. 
All inequalities among the intensity moments discussed both in the previous and this section can mutually be compared quantitatively when we transform these inequalities into the corresponding nonclassicality depths. In this approach, we replace the usual (normally-ordered) intensity moments \( \langle W^k \rangle \) by moments \( \langle W^k \rangle_s \) related to a general \( s \)-ordering of the field operators according to the formula \[3\]

\[
\langle W^k \rangle_s = \left( \frac{2}{1-s} \right)^k \left\langle \mathcal{L}_k \left( \frac{2W}{s-1} \right) \right\rangle
\]  

(83)

where \( \mathcal{L}_k \) denotes the \( k \)-th Laguerre polynomial \[37\]. Then, we formally consider all the above inequalities originally derived for the normally-ordered intensity moments with \( s \)-ordered intensity moments and varying value of the parameter \( s \). If a given inequality indicates nonclassicality for the normally-ordered moments, decreasing values of the ordering parameter \( s \) gradually suppress this nonclassicality due to the increasing additional 'detection' noise \[38\]. The nonclassicality is lost for certain threshold value \( s_{th} \). This value defines a nonclassicality depth (NCD) \( \tau \) \[38\] as follows:

\[
\tau = \frac{1-s_{th}}{2}.
\]

(84)

The greater the value of NCD \( \tau \) is the stronger the nonclassicality is.

IV. NONCLASSICALITY CRITERIA BASED ON THE ELEMENTS OF PHOTOCOUNT AND PHOTON-NUMBER DISTRIBUTIONS AND THEIR MOMENTS

All nonclassicality criteria based on intensity moments and widely discussed in the previous two sections can be easily transformed into the corresponding criteria that use the elements of photon-number [photocount] distribution \( p_{si}(n_s, n_i) \) \[30, 39–41\]. To understand this, we first write down the two-dimensional Mandel photodetection formula \[3, 4\]:

\[
p_{si}(n_s, n_i) = \frac{1}{n_s! n_i!} \int_0^\infty dW_s \int_0^\infty dW_i \left\langle W_s^{n_s} W_i^{n_i} \right\rangle \exp[-(W_s + W_i)] P_{si}(W_s, W_i),
\]

(85)

where \( P_{si}(W_s, W_i) \) is the above joint quasi-distribution of integrated intensities. Introducing the modified elements \( \tilde{p}_{si} \) of the photon-number distribution,

\[
\tilde{p}_{si}(n_s, n_i) = \frac{n_s! n_i! p_{si}(n_s, n_i)}{p_{si}(0, 0)},
\]

(86)

and the properly normalized quasi-distribution \( \tilde{P}_{si} \),

\[
\tilde{P}_{si}(W_s, W_i) = \exp[-(W_s + W_i)] P_{si}(W_s, W_i)
\times \left[ \int_0^\infty dW_s \int_0^\infty dW_i \exp[-(W_s + W_i)] P_{si}(W_s, W_i) \right]^{-1},
\]

(87)

the Mandel photodetection formula in Eq. \[85\] is recast into the form defining the modified elements \( \tilde{p}_{si} \) as the moments of the quasi-distribution \( \tilde{P}_{si} \):

\[
\tilde{p}_{si}(n_s, n_i) = \int_0^\infty dW_s \int_0^\infty dW_i W_s^{n_s} W_i^{n_i} \tilde{P}_{si}(W_s, W_i).
\]

(88)

The formal substitution in the above derived nonclassicality criteria for intensity moments suggested by formula \[85\] is expressed as

\[
\langle W_s^{n_s} W_i^{n_i} \rangle \leftarrow \tilde{p}_{si}(n_s, n_i).
\]

(89)

As an example, we rewrite the inequalities in Eq. \[19\] for \( m = 1 \) into the following Global NonClassicality Criteria:

\[
F_{kl1} \equiv \tilde{p}_{si}(k + 2, l + 2) + 2\tilde{p}_{si}(k + 1, l + 1) - 2\tilde{p}_{si}(k + 1, l + 1) < 0,
\]

\( k, l = 0, 1, \ldots \) \[90\].

Alternatively, the inequalities for intensity moments can be directly transformed into the moments of photon numbers (photocounts) exploiting the relation between the 'factorial' photon-number moments (intensity moments) \( \langle W^k \rangle \) and usual photon-number moments \( \langle n^k \rangle \). Using the Stirling numbers \( S(k, l) \) of the second kind \[28\], its two-dimensional variant is expressed in the form:

\[
\langle n^k_s n^l_i \rangle = \sum_{k_s = 1}^{k} S^{-1}(k_s, k_l) \sum_{l_i = 1}^{l} S^{-1}(k_i, l_i) W_s^{k_s} W_i^{l_i},
\]

\( k_s, k_l = 1, 2, \ldots \) \[91\].

The Stirling numbers \( S(k, l) \) of the second kind for the first five moments are conveniently expressed as a matrix \( S_{kl} \) that, together with its inverse matrix \( S_{kl}^{-1} \) giving the Stirling numbers of the first kind, take the form:

\[
S_{kl} = \begin{bmatrix}
1 & 0 & 0 & 0 & 0 \\
1 & 1 & 0 & 0 & 0 \\
1 & 3 & 1 & 0 & 0 \\
1 & 7 & 6 & 1 & 0 \\
1 & 15 & 25 & 10 & 1
\end{bmatrix},
\]

\[
S_{kl}^{-1} = \begin{bmatrix}
1 & 0 & 0 & 0 & 0 \\
-1 & 1 & 0 & 0 & 0 \\
2 & -3 & 1 & 0 & 0 \\
-6 & 11 & -6 & 1 & 0 \\
24 & -50 & 35 & -10 & 1
\end{bmatrix}.
\]

(92)

We note that the above formulas between the intensity and photon-number moments assume an effective single mode field. However, generalization to multi-mode fields may be considered, as it has been done for multi-mode twin beams in Refs. \[17, 42\]. Also, different LNNCa expressed either in the intensity or photon-number moments have been compared in \[30\].

The linear relations between the photon-number moments and the intensity moments formulated in Eq. \[91\] can be used to rewrite the nonclassicality criteria from the previous two sections in terms of the photon-number
moments. This is interesting as the joint photocount distributions are directly experimentally accessible and the joint photon-number distributions are reached once we correct the experimental data for finite detection efficiencies [43]. The rewritten nonclassicality criteria usually attain, however, more complex forms compared to the original ones written for intensity moments. For this reason, we derive here only the nonclassicality criteria that involve cross-correlation moments containing different powers of the signal and idler photon numbers. They are obtained as suitable positive linear combinations of the GNCCa \( E \) written in Eqs. (20)–(29):

\[
N_{11} \equiv E_{001} = N_{21} \equiv E_{010} + E_{011} + E_{001} = \sum_{a=s,i} [\langle n_a^2 \rangle - 2 \langle n_a \rangle - \langle n_a^2 n_i \rangle - \langle n_a n_i^2 \rangle] = 0, \tag{93}
\]

\[
N_{31} \equiv E_{020} + E_{021} + E_{111} + 3(E_{101} + E_{011} + E_{001}) = \sum_{a=s,i} [\langle n_a^3 \rangle - 3 \langle n_a^2 \rangle + 5 \langle n_a \rangle - 3 \langle n_a \rangle - 4 \langle n_a n_i \rangle - \langle n_a^2 n_i \rangle - \langle n_a n_i^2 \rangle] = 0, \tag{94}
\]

\[
N_{22} \equiv E_{201} + E_{021} + 2E_{111} + 2(E_{101} + E_{011} + E_{001}) = \sum_{a=s,i} [\langle n_a^4 \rangle - 4 \langle n_a^3 \rangle + 7 \langle n_a^2 \rangle - 4 \langle n_a \rangle - 2 \langle n_a n_i \rangle - 2 \langle n_a n_i^2 \rangle] = 0, \tag{95}
\]

\[
N_{41} \equiv E_{301} + E_{031} + E_{211} + E_{121} + 6(E_{201} + E_{021} + E_{111}) + 7(E_{101} + E_{011} + E_{001}) = \sum_{a=s,i} [\langle n_a^5 \rangle - 4 \langle n_a^4 \rangle + 6 \langle n_a^3 \rangle + 2 \langle n_a^2 \rangle - 5 \langle n_a \rangle] - 12 \langle n_a n_i \rangle - 3 \langle n_a^2 n_i \rangle - \langle n_a n_i^2 \rangle < 0, \tag{96}
\]

\[
N_{32} \equiv E_{301} + E_{031} + 2E_{211} + 2E_{121} + 4(E_{201} + E_{021}) + 7E_{111} + 4(E_{101} + E_{011}) + E_{001} = \sum_{a=s,i} [\langle n_a^4 \rangle - 6 \langle n_a^3 \rangle + 15 \langle n_a^2 \rangle - 17 \langle n_a \rangle] - 7 \langle n_a n_i \rangle - \langle n_a^2 n_i \rangle - \langle n_a n_i^2 \rangle < 0. \tag{97}
\]

\[
V. \ \text{EXPERIMENTAL VERIFICATION OF THE DERIVED NONCLASSICALITY AND ENTANGLEMENT CRITERIA}
\]

In order to experimentally judge the performance of the above derived nonclassicality criteria, we have applied them to the analysis of the entanglement between the signal and idler fields constituting a weak twin beam generated in the process of spontaneous parametric down-conversion [4, 21]. The marginal signal and idler fields are generated with multi-mode thermal statistics which is a consequence of the spontaneous emission. As such the twin beam is locally classical and so the applied GNCCa are also the entanglement criteria. The twin beam was generated in a 5-mm-long type-I barium-borate crystal (BaB\(_2\)O\(_4\), BBO) cut for a slightly non-collinear geometry (for the experimental scheme, see Fig. 1). Parametric down-conversion was pumped by pulses originating in the third harmonics (280 nm) of a femtosecond cavity dumped Ti:sapphire laser that produced pulses with duration 150 fs and central wavelength 840 nm. The signal field as well as the idler field were detected in different strips of the photocathode of iCCD camera Andor DH334-18U-63. Before detection, the nearly-frequency-degenerate signal and idler photons at the wavelength of 560 nm were filtered by a 14-nm-wide bandpass interference filter. Moreover, to stabilize the pump intensity, and thus also the twin beam intensity, to minimize fluctuations in the measured photocount distribution, the pump beam was actively stabilized via a motorized half-wave plate followed by polarizer and detector that monitored the actual intensity.

In the experiment, a joint signal-idler photocount histogram \( f_{n_1 n_2}(c_1, c_2) \) has been determined repeating the measurement \( 1.2 \times 10^6 \) times. This histogram obtained with high precision due to the high number of repetitions has allowed us to reconstruct the original joint signal-idler photon-number distribution \( p_{n_1 n_2}(n_s, n_i) \) that characterizes the twin beam before being detected. We have used two methods for making the reconstruction. First, we have applied a method developed originally for detector calibration [44]. This method, in addition of giving the detection efficiencies \( \eta_s \) and \( \eta_i \) in the signal and idler fields, respectively, also gives parameters of the used twin beam, though in a specific form of a multi-mode Gaussian field. Knowing the detection efficiencies as well as other parameters of the used iCCD camera, we have reconstructed the measured twin beam by the general approach of expectation maximization (maximum-likelihood approach) [45].

In the calibration method, the twin beam has been revealed in the analytical form of a multi-mode Gaussian field composed of independent multi-mode paired, noise signal and noise idler components characterized by mean photon-(pair) numbers \( B_a \) per mode and numbers \( M_a \) of independent modes, \( a = p, s, i \) [21, 22]. The corre-
The Mandel-Rice distribution has been theoretically analyzed in Ref. [47] and their weak noisy twin beams in multi-mode Gaussian states number distributions are multi-thermal, i.e. very classically lower than one is highly peaked around the value of parameters: $\eta_a$ and $\eta_l$ for the signal and noise idler photons. As shown below, this is manifested when considering various entanglement criteria.

We note that the POVM $T_a(c_a, n_a)$ gives the probability of having $c_a$ photocounts when detecting a field with $n_a$ photons, $a = s, i$. With these premises, the method of the least squared deviations based on the distribution $p_{si}$ in Eq. (99) and POVMs $T_a$ and $T_l$ for the signal and idler detection arm, respectively, gives both the detection efficiencies $\eta_a$ and $\eta_l$ and parameters of the used twin beam. The calibration method applied to the experimental photocount histogram $f_{si}(c_s, c_i)$ gave us the following values of parameters: $\eta_a = 0.230 \pm 0.005$, $\eta_l = 0.220 \pm 0.005$, $M_s = 270$, $B_s = 0.032$, $M_i = 0.01$, $B_i = 7.6$, $M_{si} = 0.026$, and $B_{si} = 5.3$ (relative experimental errors: 7%, for details, see [21]), in addition to those determined independently: $N_s = 6528$, $N_i = 6784$, $D_s N_s = D_i N_i = 0.040 \pm 0.001$. We note that a distribution with the number $M$ of modes considerably lower than one is highly peaked around the value $n = 0$, which is a consequence of specific form of the noise occurring in the detection process. The obtained parameters reveal that the measured weak twin beam was composed of on average 8.8 photon pairs and 0.07 (0.15) noise signal (idler) photons. Its joint signal-idler photon-number distributions $p_{si}(n_s, n_i)$ obtained by the maximum-likelihood approach as well as the calibration method, together with the experimental joint signal-idler photocount histogram $f_{si}(c_s, c_i)$ [see Fig. 2(a)], are plotted in Figs. 2(b) and 2(c), respectively. Thus, the analyzed twin beam contains tight (quantum) correlations between the signal and idler photon numbers on one side, on the other side its marginal signal and idler photon-number distributions are multi-thermal, i.e. very classical [16, 40]. We note that quantum properties of such weak noisy twin beams in multi-mode Gaussian states have been theoretically analyzed in Ref. [47] and their nonclassicality invariant describing the behavior of their entanglement on a beam-splitter has been discussed in Refs. [48, 49].

On the other hand, the application of the maximum-likelihood approach provides a joint signal-idler photon-number distribution $p_{si}(n_s, n_i)$ as a steady state of the following iteration procedure [43, 47]:

$$P_{si}^{(l+1)}(n_s, n_i) = \sum_{c_s, c_i} f_{si}(c_s, c_i) T_s(c_s, n_s) T_l(c_i, n_i) p_{si}(n_s, n_i).$$

(101)

The uniform initial distribution $p_{si}^{(0)}(n_s, n_i)$ is assumed in the iteration procedure. Compared to the joint photon-number distribution $p_{si}$ obtained in the calibration method, the distribution $p_{si}$ revealed by the iteration procedure in Eq. (101) is broader, as documented in Fig. 2(b). This reflects slightly weaker correlations between the signal and idler photon numbers (weaker pairing of photons), i.e. greater mean numbers of the noise signal and noise idler photons. As shown below, this is manifested when considering various entanglement criteria.

Nonclassicality (originating in local nonclassicality or entanglement) of a bipartite field is inscribed into its joint signal-idler quasi-distribution $P_{si}(W_s, W_i)$ of integrated intensities $W_s$ and $W_i$ that either attains negative values or even does not exist as a regular analytical function [1, 2]. In our case, we can obtain regularized forms of such quasi-distribution either by direct evaluation (for a multi-mode Gaussian field) [22] or by using the decomposition of the quasi-distribution into specific series of the Laguerre polynomials with the weights derived from the appropriate joint photocount and photon-number distributions [14]. In both cases, regularization of the quasi-distribution is provided by the experimental noise. Parallel strips with negative values are characteristic for the obtained regularized quasi-distributions $P_{si}(W_s, W_i)$ that are plotted in Fig. 3.

As the experimentally investigated noisy twin beams are mainly composed of photon pairs and exhibit multimode thermal photon-number statistics both in the signal and idler fields, they cannot be locally nonclassical, but they exhibit the entanglement. For this reason, we apply to the experimental histogram only the GNCCa derived in the previous two sections. We analyze both the joint experimental photocount histogram and the reconstructed joint photon-number distributions arising in the calibration and maximum-likelihood methods. We first pay attention to the GNCCa containing intensity moments. To allow for certain comparison among different GNCCa, we rewrite them in dimensionless units by introducing the normalized GNCCa (denoted by tildes). They are determined from the above written GNCCa by dividing them by appropriate powers of the mean intensity $\langle W \rangle = \langle W_s \rangle + \langle W_i \rangle/2$. However, fair comparison of the performance of various GNCCa containing intensity
moments of different orders is based on the corresponding (global) NCDs $\tau$ introduced in Eq. (84). In the second step and for comparison, we analyze the GNCCa given in Eqs. (93)–(98) that use photon-number moments and also some GNCCa involving the elements of photocount and photon-number distributions.

In our opinion, the GNCCa $E_{001}, \ldots, E_{121}$ given in Eqs. (20)–(29) represent the basic set of GNCCa sug-

FIG. 2. (a) Experimental photocount histogram $f_{si}(c_s, c_i)$ and reconstructed photon-number distributions $p_{si}(n_s, n_i)$ obtained by (b) maximum-likelihood and (c) calibration methods.

FIG. 3. Topo graphs of regularized quasi-distributions $P_{si}(W_s, W_i)$ of integrated intensities derived from (a) experimental photocount histogram $f_{si}$ (via its multi-mode Gaussian fit) for the ordering parameter $s = 1$, (b) photon-number distribution $p_{si}$ reconstructed by the expectation-maximization approach (via the decomposition into the Laguerre polynomials) for $s = 0$ and (c) photon-number distribution $p_{si}$ reconstructed by the calibration method (via its multi-mode Gaussian fit) for $s = 0$. In (a) [(c)], the maximum of $P_{si}$ inside the white area equals $3.6 \times 10^{-2}$ [27]. When determining $P_{si}$ in (a) and (c), one effective mode comprising the whole signal (idler) beam has been assumed [2, 15, 21, 22].
gested for the analysis of entanglement with the restriction up to the fifth-order intensity moments. This is so because of their simple forms and systematic inclusion of intensity moments of different orders. Moreover, they can be derived in parallel from the majorization theory and the inversion of simple inequalities valid for non-negative polynomials. Also, the simplest GNCC written in Eq. \( E \) was experimentally measured already in 1991 \[54\]. The values of these GNCCs determined for the experimental photocount histogram (red asterisks), reconstructed photon-number distribution using the maximum-likelihood method (green triangles) and reconstructed photon-number distribution obtained by the calibration method (blue solid curve) are plotted in Fig. 4, together with the corresponding NCDs. Except for the GNCCs \( E_{301} \) and \( E_{031} \) applied to the photocount histogram, all other GNCCs from this basic set are negative exhibiting the entanglement. Positive values of the GNCCs \( E_{301} \) and \( E_{031} \) for the photocount histogram are related to the occurrence of the fifth-order marginal intensity moments in their definitions in Eqs. (20) and (27). Both types of the applied reconstructions that partly remove the noise from the detected photocount histogram lead to negative values of the GNCCs \( E_{301} \) and \( E_{031} \). The analysis of the corresponding NCDs \( \tau \) reveals that the values of NCDs \( \tau \) decrease with the increasing order of intensity moments involved in the GNCCs. We note that similar decrease of the values of NCDs with the increasing order of intensity moments has been observed in [30] in case of LNCC. Naturally, the values of NCDs \( \tau \) are considerably greater for the reconstructed photon-number distributions compared to the original experimental photocount histogram.

The basic set of GNCC is accompanied by additional six GNCCs that are derived similarly: \( E_{002} \) [Eq. (30)], \( E_{102} \) [Eq. (31)], \( E_{012} \) [Eq. (32)], \( E_{0011} \) [Eq. (69)], \( E_{1011} \) [Eq. (65)], and \( E_{0111} \). Unfortunately, none of these GNCCs indicates the entanglement in the measured twin beam, as documented in Fig. 5. Positive values of the GNCCs \( E_{002} \), \( E_{102} \) and \( E_{012} \) can again be related to the presence of the fourth- and fifth-order marginal intensity moments in the definitions of these GNCCs. On the other hand, the GNCCs \( E_{0011} \), \( E_{1011} \) and \( E_{0111} \) contain in their definitions the terms with two and even three intensity moments in a product, which seriously limits their ability to reveal entanglement.

Restricting our consideration to the fourth-order intensity moments, the majorization theory provides five GNCCs [denoted by symbol \( D \), Eqs. (17)–(19)] for which products of two intensity moments are characteristic, together with nine GNCCs [denoted by symbol \( T \), Eqs. (50)–(55)] containing terms with up to three intensity moments in a product. All these GNCCs indicate by their negative values the entanglement both in the photocount histogram and the reconstructed photon-number distributions, as documented in Figs. 6 and 7. Mutual comparison of NCDs \( \tau \) for the GNCCs \( E \), \( D \) and \( T \) plotted in turn in Figs. 4, 6 and 7 reveals that the entanglement described by the GNCC \( E \) is the most resistant against the noise, the GNCC \( D \) are considerably worse from this point of view and the resistance of the GNCC \( T \) against the noise is already weak. This behavior can qualitatively be explained by the occurrence of multiple products of intensity moments in the expressions giving the GNCC \( D \) and \( T \). These products do not naturally describe any correlation and so their presence in the GNCC only weakens the ability of a given GNCC to identify the entanglement.

The widely used matrix approach [19, 23, 24] gives us three GNCC \( M_{1100} \) [Eq. (73)], \( M_{1001} \) [Eq. (74)] and \( M_{00101} \) [Eq. (75)] for investigating entanglement, provided that intensity moments up to the fifth order are taken into account. For our experimental data, only the GNCC \( M_{1001} \) and \( M_{00101} \) identify entanglement (see Fig. 8). We note that negativity of the experimental GNCC \( M_{1001} \) has been reported in [17]. The values of the corresponding NCDs \( \tau \) plotted in Fig. 8 are comparable to those characterizing the GNCC \( E \) from the basic set. This shows their high performance in identifying the entanglement. A bit surprisingly the GNCC \( M_{1100} \) is positive. In our opinion this is a consequence
of the thermal statistics of photon pairs. Loosely speaking and relying on the quantum theory, we may define ‘a photon-pair intensity’\( W_{si} \approx W_s W_i \) that allows us to rewrite Eq. (73) in the form \( M_{1100} \approx \langle W_{si}^2 \rangle - \langle W_{si} \rangle^2 \) that explains positivity of the GNCC \( M_{1100} \) for the analyzed weak twin beam.

The Cauchy-Schwarz inequality provides two simple GNCCs not mentioned above, \( C_{12}^{10} \) [Eq. (80) and \( C_{01}^{21} \) [Eq. (82)] whose performance in revealing the entanglement lies in between the GNCCs \( M_{1001} \) and \( M_{1100} \) (see Fig. 5). For the experimental twin beam, only the GNCC \( C_{01}^{21} \) applied to the reconstructed photon-number distributions indicates the entanglement. As the GNCC \( C_{12}^{10} \) is derived from the GNCC \( C_{01}^{21} \) by substitution \( s \leftrightarrow i \), this demonstrates strong sensitivity of both GNCCs to the level of noise. The slightly lower mean of the signal noise photon number compared to that of the idler field (0.07 versus 0.15) is sufficient to observe the negative GNCC \( C_{01}^{21} \). For comparison, we plot in Fig. 8 another two GNCCs \( D_{1111}^{2100} \) [Eq. (75)] and \( D_{1111}^{2100} \) [Eq. (76)] that also contain the cross-correlation intensity moments \( \langle W_s W_i \rangle \) and \( \langle W_s^2 W_i^2 \rangle \) and that are expressed as positive linear combinations of the already analyzed GNCCs. However, their NCDs \( \tau \) are lower due to the additional terms with marginal higher-order intensity moments occurring in their definitions compared to the formulas for the GNCCs \( M \) written in Eqs. (74) and (77).

All the above discussed GNCCs that are based on the intensity moments can straightforwardly be converted into the corresponding GNCCs that contain photocount and photon-number moments using the linear relations between both types of moments quantified by the Stirling numbers \( S \) [see Eq. (92)]. This is more-or-less formal for the reconstructed photon-number distributions. Contrary to this, such GNCCs are useful and convenient when experimental photocount histograms are analyzed. The reason is that these GNCCs can directly be applied to the experimental data. This is why we have suitably combined together various GNCCs written for the intensity moments to arrive at a specific set of six simple GNCCs \( N \) written in Eqs. (93)—(98). All of them have been able to reveal the entanglement in the experimental histogram, as documented in Fig. 9. However, we note that the GNCCs \( N \) are expressed as sums of intensity moments of different orders and, as such, their structure is less transparent compared to the original GNCCs based on the intensity moments.

The comparison of the results reached by the above discussed GNCCs applied to the photon-number distributions reconstructed by the maximum-likelihood approach and the calibration method reveals the following. Negative values of the GNCCs, that reveal the entanglement, reached by both approaches equal within the experimental errors or the values provided by the maximum-
FIG. 8. (a) Normalized global nonclassicality criteria \( \tilde{M}, \tilde{C}, \tilde{D} \) defined in Eqs (73)–(77), (80), and (82) and (b) the corresponding nonclassicality depths \( \tau \). For description, see the caption to Fig. 4.

FIG. 9. Normalized global nonclassicality criteria \( \tilde{N} \) defined in Eqs (93)–(98). For description, see the caption to Fig. 4. Normalization is done with respect to the corresponding quantities \( N_{\text{ref}} \) determined for the factorized distribution \( P_s(W_s)P_i(W_i) \).

likelihood approach are greater than those reached by the calibration method. In consequence, the corresponding NCDs from both approaches coincide within the experimental errors or those arising in the calibration method are greater. This behavior naturally stems from the fact that the calibration method is more efficient in removing the noise from the experimental data. This is so as the calibration method works with a pre-defined form of the photon-number distribution and applies it simultaneously to the whole 2D experimental photocount histogram.

Finally, all the above written GNCCa as well as LNCCa can be transformed into the corresponding GNCCa and LNCCa that involve the elements of photocount histogram or reconstructed photon-number distributions using the formal substitution written in Eq. (89). The use of such GNCCa, however, needs different approach compared to that applied to the GNCCa containing intensity moments. Whereas only the intensity moments up to certain order are useful owing to the increasing experimental error with the increasing order of intensity moment, useful and reliable GNCCa in case of the distributions involve their elements (probabilities) having the highest available values. As both the joint photocount histogram \( f_{s1} \) and the joint reconstructed photon-number distributions \( p_{s1} \) have such elements around the diagonal (see Fig. 4), we consider the GNCCa involving the elements at the diagonal \( \{41, 51\} \) and the closest neighbor parallel lines, as described in turn by functions \( F_{kk}, F_{(k+j)k1} \) and \( F_{k(k+j)1} \), \( j = 1, 2 \), with the varying index \( k \) (see Fig. 10). The GNCCa \( F \) defined in Eq. (90) reveal reliably the entanglement via their negative values in the area around the peaks of both the photocount histogram \( (k \approx 2) \) and reconstructed photon-number distributions \( (k \approx 9) \). We note that negative values of the GNCCa \( F_{(k+j)k1} \) and \( F_{k(k+j)1} \) for \( j = 2, \ldots, \{j = 1, \ldots\} \) have not been observed for the photon-number distribution reconstructed by the maximum-likelihood [calibration] method which is a consequence of its narrow ‘cigar’ shape clearly visible in Fig. 2(b) [2(c)].

VI. CONCLUSIONS

We have derived numerous inequalities among the moments of integrated intensities aimed at identifying local as well as global nonclassicality using a) the majorization theory, b) non-negative polynomials, c) the matrix approach based on quadratic forms and d) the Cauchy-Schwarz inequality. We have mutually compared different approaches, grouped the obtained nonclassicality criteria according to their structure and tested their performance on the experimental data characterizing a weak twin beam with about nine photon pairs per pulse and small amount of an additional noise. We have identified a basic set of ten global nonclassicality criteria, that have revealed the entanglement in the analyzed twin beam. We have also paid attention to the counterparts of nonclassicality criteria written in the moments of photocounts and photon numbers and also the elements of photocount and photon-number distributions. We have demonstrated their performance on the same experimental data. For twin beams with low amount of the noise all three different kinds of nonclassicality criteria represent a strong tool for revealing the entanglement.
FIG. 10. Normalized global nonclassicality criteria $\tilde{F}_{k^l}$ given in Eq. (9) for (a) experimental photocount histogram $f_{ai}$ and $k' = kk$ (red asterisks), $(k + 1)k$ (green), $(k + 2)k$ (yellow), $k(k + 1)$ (light blue), $k(k + 2)$ (dark blue), (b) photon-number distribution $p_{ai}$ reconstructed by the maximum-likelihood approach and $k' = kk$ (red triangles), $(k + 1)k$ (green), $k(k + 1)$ (blue) and (c) photon-number distribution $p_{ai}$ reconstructed by the calibration method for $k' = kk$ (solid blue curve); $\tilde{F}_{k^l} = [(k + 1)(k + 2)p_{ai}(k + 2, l) + (l + 1)(l + 2)p_{ai}(k, l + 2) - 2(k + 1)(l + 1)p_{ai}(k + 1, l + 1)] / [(k + 1)(k + 2)p_{ai}(k + 2, l) + (l + 1)(l + 2)p_{ai}(k, l + 2) + (l + 1)p_{ai}(k, l + 1)]$ and $p_{ai}(n)$ is the Poissonian distribution with mean $\langle n_{ai} \rangle$ normalized such that $p_{ai}^{\nu}(0) = 1$, $a = s, i$.

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Appendix A: Additional (redundant) nonclassicality criteria

In Appendix A, we summarize the nonclassicality criteria derived from the majorization theory with polynomials written in three and four variables and being redundant with respect to those presented in the main text. This means that such LNCCa and GNCCa are expressed as positive linear combinations of the LNCCa and GNCCa written in the main text.

First, we summarize the redundant (and properly normalized) LNCCa that complement the LNCCa contained in Eqs. (35)–(40) and (56)–(59) ($a = s, i$):

\begin{align}
{a}B_{110}^{200} &= aL_{11}^{20} + B_{11}^{20} < 0, \\
{a}B_{210}^{300} &= aL_{21}^{30} + B_{21}^{30} < 0, \\
{a}B_{310}^{400} &= aL_{31}^{40} + B_{31}^{40} < 0, \\
{a}B_{220}^{320} &= aL_{22}^{32} + B_{22}^{32} < 0, \\
{a}B_{210}^{420} &= 2aL_{21}^{42} + B_{21}^{42} < 0, \\
{a}B_{110}^{520} &= sL_{11}^{20} + sL_{11}^{20} + 2B_{11}^{20} < 0, \\
{a}B_{210}^{330} &= 2aL_{21}^{33} + B_{21}^{33} < 0, \\
{a}B_{310}^{440} &= sL_{31}^{44} + sL_{31}^{44} + 2B_{31}^{44} < 0, \\
{a}B_{220}^{322} &= 2aL_{22}^{322} + B_{22}^{322} < 0, \\
{a}B_{110}^{522} &= sL_{11}^{22} + sL_{11}^{22} + 2B_{11}^{22} < 0, \\
{a}B_{210}^{332} &= 2aL_{21}^{332} + B_{21}^{332} < 0, \\
{a}B_{310}^{442} &= sL_{31}^{442} + sL_{31}^{442} + 2B_{31}^{442} < 0, \\
{a}B_{220}^{322} &= 2aL_{22}^{322} + B_{22}^{322} < 0, \\
{a}B_{110}^{522} &= sL_{11}^{22} + sL_{11}^{22} + 2B_{11}^{22} < 0, \\
{a}B_{210}^{332} &= 2aL_{21}^{332} + B_{21}^{332} < 0, \\
{a}B_{310}^{442} &= sL_{31}^{442} + sL_{31}^{442} + 2B_{31}^{442} < 0, \\
{a}B_{220}^{322} &= 2aL_{22}^{322} + B_{22}^{322} < 0, \\
{a}B_{110}^{522} &= sL_{11}^{22} + sL_{11}^{22} + 2B_{11}^{22} < 0. \\
\end{align}

The redundant (and properly normalized) GNCCa containing the terms with up to two intensity moments in a product attain the form ($a = s, i$):

\begin{align}
{a}D_{110}^{200} &= aL_{11}^{20} + (E_{001} + B_{11}^{20})/2 < 0, \\
{a}D_{210}^{300} &= 2aL_{21}^{30} + E_{011} + E_{011} + B_{21}^{30} < 0, \\
{a}D_{310}^{400} &= 2aL_{31}^{40} + E_{021} + E_{021} + B_{31}^{40} < 0, \\
{a}D_{220}^{320} &= 2aL_{22}^{32} + E_{021} + E_{021} + B_{22}^{32} < 0, \\
{a}D_{210}^{420} &= 2aL_{21}^{42} + E_{011} + E_{011} + B_{21}^{42} < 0, \\
{a}D_{310}^{440} &= 2aL_{31}^{44} + E_{011} + E_{011} + B_{31}^{44} < 0, \\
{a}D_{220}^{322} &= 2aL_{22}^{322} + E_{021} + E_{021} + B_{22}^{322} < 0, \\
{a}D_{310}^{442} &= 2aL_{31}^{442} + E_{011} + E_{011} + B_{31}^{442} < 0, \\
{a}D_{220}^{322} &= 2aL_{22}^{322} + E_{021} + E_{021} + B_{22}^{322} < 0, \\
{a}D_{110}^{520} &= 2aL_{11}^{52} + E_{011} + B_{11}^{52} < 0, \\
{a}D_{110}^{522} &= 2aL_{11}^{522} + E_{011} + B_{11}^{522} < 0, \\
\end{align}

Finally, the redundant (and properly normalized) GNCCa expressed via triple products of intensity moments are derived as follows ($a = s, i$):

\begin{align}
{a}T_{1100}^{200} &= 6aL_{11}^{20} + E_{001} + 2B_{11}^{20} < 0, \\
\end{align}
\begin{align}
T_{1100}^{2000} &= \frac{s}{2} L_{21}^{30} + \frac{i}{2} L_{11}^{20} + (E_{001} + 3B_{21}^{20})/2 < 0, \quad (A28) \\
\alpha T_{1100}^{3000} &= 6 \frac{s}{2} L_{21}^{30} + E_{101} + E_{011} + 2B_{21}^{20} < 0, \quad (A29) \\
T_{2100}^{3000} &= 2 \frac{s}{2} L_{21}^{30} + 2 \frac{i}{2} L_{21}^{30} + E_{101} + E_{011} + 3B_{21}^{20} < 0, \quad (A30) \\
\alpha T_{3100}^{4000} &= 6 \frac{s}{2} L_{31}^{40} + E_{201} + E_{111} + E_{021} + 2B_{31}^{40} < 0, \quad (A31) \\
T_{3100}^{4000} &= 2 \frac{s}{2} L_{31}^{40} + 2 \frac{i}{2} L_{31}^{40} + E_{201} + E_{111} + E_{021} + 3B_{31}^{40} < 0, \quad (A32) \\
\alpha T_{2200}^{3100} &= 6 \frac{s}{2} L_{22}^{31} + E_{111} + 2B_{22}^{31} < 0, \quad (A33) \\
T_{2200}^{3100} &= 2 \frac{s}{2} L_{22}^{31} + 2 \frac{i}{2} L_{22}^{31} + E_{111} + 3B_{22}^{31} < 0. \quad (A34)
\end{align}

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