Testing nonclassicality in multimode fields: A unified derivation of classical inequalities

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We consider a way to generate operational inequalities to test nonclassicality (or quantumness) of multimode bosonic fields (or multiparty bosonic systems) that unifies the derivation of many known inequalities and allows to propose new ones. The nonclassicality criteria are based on Vogel's criterion corresponding to analyzing the positivity of multimode P functions or, equivalently, the positivity of matrices of expectation values of, e.g., creation and annihilation operators. We analyze not only monomials but also polynomial functions of such moments, which can sometimes enable simpler derivations of physically relevant inequalities. As an example, we derive various classical inequalities which can be violated only by nonclassical fields. In particular, we show how the criteria introduced here easily reduce to the well-known inequalities describing (a) multimode quadrature squeezing and its generalizations, including sum, difference, and principal squeezing; (b) two-mode one-time photon-number correlations, including sub-Poisson photon-number correlations and effects corresponding to violations of the Cauchy-Schwarz and Muirhead inequalities; (c) two-time single-mode photon-number correlations, including photon antibunching and hyperbunching; and (d) two- and three-mode quantum entanglement. Other simple inequalities for testing nonclassicality are also proposed. We have found some general relations between the nonclassicality and entanglement criteria, in particular those resulting from the Cauchy-Schwarz inequality. It is shown that some known entanglement inequalities can be derived as nonclassicality inequalities within our formalism, while some other known entanglement inequalities can be seen as sums of more than one inequality derived from the nonclassicality criterion. This approach enables a deeper analysis of the entanglement for a given nonclassicality.

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I. INTRODUCTION

Testing whether a given state of a system cannot be described within a classical theory has been one of the fundamental problems of quantum theory from its beginnings to current studies in, e.g., quantum optics [1–12], condensed matter (see, e.g., Refs. [3,13]), nanomechanics [14,15], and quantum biology (see, e.g., Ref. [16]). Macroscopic quantum superpositions (being at the heart of the Schrödinger-cat paradox) and related entangled states (which are at the core of the Einstein-Podolsky-Rosen paradox and Bell's theorem) are famous examples of nonclassical states which are not only physical curiosities but now fundamental resources for quantum-information processing [17].

All states (or phenomena) are quantum, i.e., nonclassical. Thus, it is quite arbitrary to call some states "classical." Nevertheless, some states are closer to their classical approximation than other states. The most classical *pure* states of the harmonic oscillator are coherent states. Thus, usually, they are considered classical, while all other pure states of the harmonic oscillator are deemed nonclassical. The nonclassicality criterion for *mixed* states is more complicated and it is based on the Glauber-Sudarshan P function [1,2]. A commonly accepted formal criterion which enables to distinguish nonclassical from classical states reads as follows [3–6]: A quantum state is *nonclassical* if its Glauber-Sudarshan P function cannot be interpreted as a *true* probability density. Note that, according to this definition, any entangled state is nonclassical, but not every separable state is classical.

Various operational criteria of nonclassicality (or quantumness) of single-mode fields were proposed (see, e.g., Refs. [3,4,18,19] and references therein). In particular, Agarwal and Tara [20] and Shchukin, Richter, and Vogel (SRV) [21,22] proposed nonclassicality criteria based on matrices of moments of annihilation and creation operators for single-mode fields. Moreover, an efficient method for measuring such moments was also developed by Shchukin and Vogel [23].

It is not always sufficient to analyze a single-mode field, i.e., an elementary excitation of a normal mode of the field confined to a one-dimensional cavity. To describe the generation or interaction of two or more bosonic fields, the standard analysis of single-system nonclassicality should be generalized to the two- and multisystem (multimode) cases. Simple examples of such bosonic fields are multimode number states, multimode coherent and squeezed light, or fields generated in multiwave mixing, multimode scattering, or multiphoton resonance.

Here, we study in greater detail and modify an operational criterion of nonclassicality for multimode radiation fields of Vogel [24], which is a generalized version of the SRV nonclassicality criterion [21,22] for single-mode fields. It not only describes the multimode fields but can also be applied in the analysis of the dynamics of radiation sources. This could be important for the study of, e.g., time-dependent correlation functions, which are related to time-dependent field commutation rules (see, e.g., subsections 2.7 and 2.8 in Ref. [4]).

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A variety of multimode nonclassicality inequalities has been proposed in quantum optics (see, e.g., textbooks [3–6], reviews [7–11], and Refs. [25–41]) and tested experimentally (see, e.g., Refs. [42–48]). The nonclassicality criterion described here enables a simple derivation of them. Moreover, it offers an effective way to derive new inequalities, which might be useful in testing the nonclassicality of specific states generated in experiments. It is worth noting that we are analyzing nonclassicality criteria but *not* a degree of nonclassicality. We admit that the latter problem is experimentally important and a few "measures" of nonclassicality have been proposed [49–58].

Analogously to the SRV nonclassicality criteria, Shchukin and Vogel [59] proposed an entanglement criterion based on the matrices of moments and partial transposition. This criterion was later amended [60] and generalized [61] to replace partial transposition by nondecomposable positive maps and contraction maps (e.g., realignment). A similar approach for entanglement verification, based on the construction of matrices of expectation values, was also investigated in Refs. [62–65]. Here we analyze relations between classical inequalities derived from the two- and three-mode nonclassicality criteria and the above-mentioned entanglement criterion.

The article is organized as follows: In Sec. II, a nonclassicality criterion for multimode bosonic fields is formulated. We apply the criterion to rederive known and a few apparently new nonclassicality inequalities. In subsection III A, we summarize the Shchukin-Vogel entanglement criterion [59,60]. In subsection III C, we apply it to show that some known entanglement inequalities (including those of Duan *et al.* [66] and Hillery and Zubairy [67]) exactly correspond to unique nonclassicality inequalities. In subsection III D, we analyze such entanglement inequalities (including Simon's criterion [68]) that are represented apparently not by a single inequality but by sums of inequalities derived from the nonclassicality criterion. Moreover, other entanglement inequalities are derived in subsection III D2. The discussed nonclassicality and entanglement criteria are summarized in Tables I and II. We conclude in Sec. IV.

II. NONCLASSICALITY CRITERIA FOR MULTIMODE FIELDS

An M-mode bosonic state $\hat{\rho}$ can be completely described by the Glauber-Sudarshan P function defined by [1,2]:

$$\hat{\rho} = \int d^2\boldsymbol{\alpha}\; P(\boldsymbol{\alpha},\boldsymbol{\alpha}^*)|\boldsymbol{\alpha}\rangle\langle\boldsymbol{\alpha}|, \tag{1}$$

where $|\boldsymbol{\alpha}\rangle = \prod_{m=1}^{M} |\alpha_m\rangle$ and $|\alpha_m\rangle$ is the mth-mode coherent state, i.e., the eigenstate of the mth-mode annihilation operator $\hat{a}_m$, $\boldsymbol{\alpha}$ denotes complex multivariable $(\alpha_1,\alpha_2,\ldots,\alpha_M)$, and $d^2\boldsymbol{\alpha} = \prod_m d^2\alpha_m$. The density matrix $\hat{\rho}$ can be supported on the tensor product of either infinite-dimensional or finite-dimensional Hilbert spaces. For the sake of simplicity, we assume the number M of modes to be finite. But there is no problem to generalize our results for an infinite number of modes.

A criterion of nonclassicality is usually formulated as follows [70]:

Criterion 1. A multimode bosonic state $\hat{\rho}$ is considered to be nonclassical if its Glauber-Sudarshan P function cannot be interpreted as a classical probability density, i.e., it is nonpositive or more singular than Dirac's δ function. Conversely, a state is called classical if it is described by a P function being a classical probability density.

It is worth noting that Criterion 1 (and the following criteria) does not have a fundamental indisputable validity, and it was the subject of criticism by, e.g., Wünsche [71], who made the following two observations. (i) In the vicinity of any classical state there are nonclassical states, as can be illustrated by analyzing modified thermal states. So, arbitrarily close to any classical state there is a nonclassical state giving, to arbitrary precision, exactly the same outcomes as for the classical state in any measurement. Note that analogous problems can be raised for entanglement criteria [61] for continuous-variable systems, as in the vicinity of any separable state there are entangled states.[1] (ii) There are intermediate quasiclassical (or unorthodox classical) states, which *cannot* be clearly classified as classical or nonclassical according to Criterion 1. This can be illustrated by analyzing the squeezing of thermal states, which does not lead immediately from classical to nonclassical states. Due to the singularity of the P function, Criterion 1 is not operationally useful as it is extremely difficult (although sometimes possible [72]) to directly reconstruct the P function from experimental data.

Recently, Shchukin, Richter, and Vogel [21,22] proposed a hierarchy of operational criteria of nonclassicality of single-mode bosonic states. This approach is based on the normally ordered moments of, e.g., annihilation and creation operators or position and momentum operators. An infinite set of these criteria (by inclusion of the correction analogous to that given in Ref. [60]) corresponds to a single-mode version of Criterion 1.

Let us consider a (possibly infinite) countable set $\hat{F} = (\hat{f}_1,\hat{f}_2,\ldots,\hat{f}_i,\ldots)$ of M-mode operators $\hat{f}_i \equiv \hat{f}_i(\hat{\mathbf{a}},\hat{\mathbf{a}}^\dagger)$, each a function of annihilation, $\hat{\mathbf{a}} \equiv (\hat{a}_1,\hat{a}_2,\ldots,\hat{a}_M)$, and creation, $\hat{\mathbf{a}}^\dagger$, operators. For example, we may choose such operators as monomials

$$\hat{f}_i = \prod_{m=1}^{M} (\hat{a}_m^\dagger)^{i_{2m-1}} \hat{a}_m^{i_{2m}}, \tag{2}$$

where i stands in this case for the multi-index $\mathbf{i} \equiv (i_1,i_2,\ldots,i_{2M})$, but the $\hat{f}_i$'s can be more complicated functions, for example, polynomials in the creation and annihilation operators.

If

$$\hat{f} = \sum_i c_i \hat{f}_i, \tag{3}$$

where c_i are arbitrary complex numbers, then with the help of the P function one can directly calculate the normally ordered (denoted by ::) mean values of the Hermitian operator $\hat{f}^\dagger\hat{f}$ as follows [21,73]:

$$\langle{:}\hat{f}^\dagger\hat{f}{:}\rangle = \int d^2\boldsymbol{\alpha}\; |f(\boldsymbol{\alpha},\boldsymbol{\alpha}^*)|^2 P(\boldsymbol{\alpha},\boldsymbol{\alpha}^*). \tag{4}$$

[1] It is worth stressing that this is the case only for continuous-variable systems: in the finite dimensional case, the set of separable states has finite volume.

TABLE I. Criteria for single-time nonclassical effects in two-mode (TM) and multimode (MM) fields, and two-time nonclassical effects in single-mode (SM) fields.

Nonclassical effect	Criterion	Equations
MM quadrature squeezing	$d^{(n)}(1,\hat{X}_\phi) < 0$	(A1), (A6)
TM principal squeezing of Lukš *et al.* [32]	$d^{(n)}(\Delta\hat{a}_{12}^\dagger,\Delta\hat{a}_{12}) = d^{(n)}(1,\hat{a}_{12}^\dagger,\hat{a}_{12}) < 0$	(A7)–(A10)
TM sum squeezing of Hillery [33]	$d^{(n)}(1,\hat{V}_\phi) < 0$	(A12), (A15)
MM sum squeezing of An-Tinh [39]	$d^{(n)}(1,\hat{\mathcal{V}}_\phi) < 0$	(A18), (A20)
TM difference squeezing of Hillery [33]	$d^{(n)}(1,\hat{W}_\phi) < -\frac{1}{2}\min(\langle\hat{n}_1\rangle,\langle\hat{n}_2\rangle)$	(A21), (A25), (A26)
MM difference squeezing of An-Tinh [40]	$d^{(n)}(1,\hat{\mathcal{W}}_\phi) < -\frac{1}{4}\|\|\langle\hat{C}\rangle\| - \langle\hat{D}\rangle\|$	(A31), (A34)
TM sub-Poisson photon-number correlations	$d^{(n)}(1,\hat{n}_1 \pm \hat{n}_2) < 0$	(B1), (B3)
Cauchy-Schwarz inequality violation	$d^{(n)}(\hat{f}_1,\hat{f}_2) < 0$	(15), (16)
TM Cauchy-Schwarz inequality violation via Agarwal's test [31]	$d^{(n)}(\hat{n}_1,\hat{n}_2) < 0$	(B4), (B6)
TM Muirhead inequality violation via Lee's test [34]	$d^{(n)}(\hat{n}_1 - \hat{n}_2) < 0$	(B7), (B8)
SM photon antibunching	$d^{(n)}[\hat{n}(t),\hat{n}(t+\tau)] < 0$	(C3), (C6)
SM photon hyperbunching	$d^{(n)}[\Delta\hat{n}(t),\Delta\hat{n}(t+\tau)]$	(C7), (C12), (C13)
	$= d^{(n)}[1,\hat{n}(t),\hat{n}(t+\tau)] < 0$	
Other TM nonclassical effects	$d^{(n)}(1,\hat{a}_1\hat{a}_2,\hat{a}_1^\dagger\hat{a}_2^\dagger) < 0$	(18)
	$d^{(n)}(1,\hat{a}_1\hat{a}_2^\dagger,\hat{a}_1^\dagger\hat{a}_2) < 0$	(19)
	$d^{(n)}(1,\hat{a}_1+\hat{a}_2^\dagger,\hat{a}_1^\dagger+\hat{a}_2) < 0$	(20)
	$d^{(n)}(1,\hat{a}_1+\hat{a}_2,\hat{a}_1^\dagger+\hat{a}_2^\dagger) < 0$	(21)
	$d^{(n)}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger,\hat{a}_2) < 0$	(22)

The crucial observation of SRV [21] in the derivation of their criterion is the following:

Observation 1. If the P function for a given state is a classical probability density, then $\langle{:}\hat{f}^\dagger\hat{f}{:}\rangle \geqslant 0$ for any function $\hat{f}$. Conversely, if $\langle{:}\hat{f}^\dagger\hat{f}{:}\rangle < 0$ for some $\hat{f}$, then the P function is not a classical probability density.

The condition based on nonpositivity of the P function is usually considered a necessary and sufficient condition of nonclassicality. In fact, as shown by Sperling [74], if the P function is more singular than Dirac's δ function [e.g., given by the nth derivative of $\delta(\alpha)$ for $n = 1,2,\ldots$], then it is also nonpositive.

TABLE II. Entanglement criteria via nonclassicality criteria.

Reference	Entanglement criterion	Equivalent nonclassicality criterion	Equations
Duan *et al.* [66]	$d^\Gamma(\Delta\hat{a}_1,\Delta\hat{a}_2) = d^\Gamma(1,\hat{a}_1,\hat{a}_2) < 0$	$d^{(n)}(\Delta\hat{a}_1,\Delta\hat{a}_2^\dagger) = d^{(n)}(1,\hat{a}_1,\hat{a}_2^\dagger) < 0$	(48)–(50)
Simon [68]	$d^\Gamma(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2,\hat{a}_2^\dagger) < 0$	$d^{(n)}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger,\hat{a}_2) + d^{(n)}(1,\hat{a}_1,\hat{a}_2^\dagger)$	(52)
		$+d^{(n)}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger) + d^{(n)}(1,\hat{a}_1,\hat{a}_2^\dagger,\hat{a}_2) < 0$	
Mancini *et al.* [69]	$d^\Gamma(1,\hat{a}_1+\hat{a}_2,\hat{a}_1^\dagger+\hat{a}_2^\dagger) < 0$	$d^{(n)}(1,\hat{a}_1+\hat{a}_2^\dagger,\hat{a}_1^\dagger+\hat{a}_2) + 2d^{(n)}(1,\hat{a}_1+\hat{a}_2^\dagger) + 1 < 0$	(58), (59)
Hillery & Zubairy [67]	$d^\Gamma(1,\hat{a}_1\hat{a}_2) < 0$	$d^{(n)}(1,\hat{a}_1\hat{a}_2^\dagger) < 0$	(33), (36)
Hillery & Zubairy [67]	$d^\Gamma(1,\hat{a}_1^m\hat{a}_2^n) < 0$	$d^{(n)}(1,\hat{a}_1^m(\hat{a}_2^\dagger)^n) < 0$	(40)–(42)
Hillery & Zubairy [67]	$d^\Gamma(\hat{a}_1,\hat{a}_2) < 0$	$d^{(n)}(\hat{a}_1,\hat{a}_2^\dagger) < 0$	(34), (37)
Hillery & Zubairy [67]	$d^\Gamma(1,\hat{a}_1\hat{a}_2\hat{a}_3) < 0$	$d^{(n)}(1,\hat{a}_1^\dagger\hat{a}_2\hat{a}_3) < 0$	(35), (38)
Miranowicz *et al.* [61]	$d^\Gamma(\hat{a}_1,\hat{a}_2\hat{a}_3) < 0$	$d^{(n)}(\hat{a}_1^\dagger,\hat{a}_2\hat{a}_3) < 0$	(39)
Other entanglement tests	$d^\Gamma(1,\hat{a}_1^k\hat{a}_2^l\hat{a}_3^m) < 0$	$d^{(n)}(1,(\hat{a}_1^\dagger)^k\hat{a}_2^l\hat{a}_3^m) < 0$	(43), (44)
	$d^\Gamma(\hat{a}_1^k,\hat{a}_2^l\hat{a}_3^m) < 0$	$d^{(n)}((\hat{a}_1^\dagger)^k,\hat{a}_2^l\hat{a}_3^m) < 0$	(45), (46)
	$d^\Gamma(1,\hat{a}_1\hat{a}_2,\hat{a}_1^\dagger\hat{a}_2^\dagger) < 0$	$d^{(n)}(1,\hat{a}_1\hat{a}_2^\dagger,\hat{a}_1^\dagger\hat{a}_2) + (\langle\hat{n}_1+\hat{n}_2\rangle+1)d^{(n)}(1,\hat{a}_1\hat{a}_2^\dagger) < 0$	(54), (55)
	$d^\Gamma(1,\hat{a}_1\hat{a}_2^\dagger,\hat{a}_1^\dagger\hat{a}_2) < 0$	$d^{(n)}(1,\hat{a}_1\hat{a}_2,\hat{a}_1^\dagger\hat{a}_2^\dagger) + \langle\hat{n}_1\rangle\langle\hat{n}_2\rangle + \langle\hat{n}_1+\hat{n}_2\rangle d^{(n)}(1,\hat{a}_1\hat{a}_2) < 0$	(56), (57)
	$d^\Gamma(1,\hat{a}_1+\hat{a}_2,\hat{a}_1^\dagger+\hat{a}_2^\dagger) < 0$	$d^{(n)}(1,\hat{a}_1+\hat{a}_2^\dagger,\hat{a}_1^\dagger+\hat{a}_2) + 2d^{(n)}(1,\hat{a}_1+\hat{a}_2^\dagger) < 0$	(60), (61)

With the help of Eq. (3), $\langle :\hat{f}^\dagger \hat{f}: \rangle$ can be given by

$$\langle :\hat{f}^\dagger \hat{f}: \rangle = \sum_{i,j} c_i^* c_j M_{ij}^{(\mathrm{n})}(\hat{\rho}) \tag{5}$$

in terms of the normally ordered correlation functions

$$M_{ij}^{(\mathrm{n})}(\hat{\rho}) = \mathrm{Tr}(:\hat{f}_i^\dagger \hat{f}_j : \hat{\rho}), \tag{6}$$

where the superscript (n) (similarly to ::) denotes the normal order of field operators. In the special case of two modes, analyzed in detail in the next sections, and with the choice of $\hat{f}_i$ given by Eq. (2), Eq. (6) can be simply written as

$$M_{ij}^{(\mathrm{n})}(\hat{\rho}) = \mathrm{Tr}[:(\hat{a}^{\dagger i_1}\hat{a}^{i_2}\hat{b}^{\dagger i_3}\hat{b}^{i_4})^\dagger(\hat{a}^{\dagger j_1}\hat{a}^{j_2}\hat{b}^{\dagger j_3}\hat{b}^{j_4}) : \hat{\rho}], \tag{7}$$

where $\hat{a} = \hat{a}_1$ and $\hat{b} = \hat{a}_2$. It is worth noting that there is an efficient optical scheme [23] for measuring the correlation functions (7).

With a set $\hat{F} = (\hat{f}_1, \hat{f}_2, \ldots, \hat{f}_i, \ldots)$ fixed, the correlations (6) form a (possibly infinite) Hermitian matrix

$$M^{(\mathrm{n})}(\hat{\rho}) = \left[M_{ij}^{(\mathrm{n})}(\hat{\rho})\right]. \tag{8}$$

In order to emphasize the dependence of (8) on the choice of $\hat{F}$, we may write $M_{\hat{F}}^{(\mathrm{n})}(\hat{\rho})$. Moreover, let $[M^{(\mathrm{n})}(\hat{\rho})]_{\mathbf{r}}$, with $\mathbf{r} = (r_1, \ldots, r_N)$, denote the $N \times N$ principal submatrix of $M^{(\mathrm{n})}(\hat{\rho})$ obtained by deleting all rows and columns except the ones labeled by $r_1, \ldots, r_N$.

In analogy to Vogel's approach [24], by applying Sylvester's criterion (see, e.g., Refs. [60,75]) to the matrix (8), a generalization of the single-mode SRV criterion for multimode fields can be formulated as follows:

Criterion 2. For any choice of $\hat{F} = (\hat{f}_1, \hat{f}_2, \ldots, \hat{f}_i, \ldots)$, a multimode state $\hat{\rho}$ is nonclassical if there exists a negative principal minor, i.e., $\det(M_{\hat{F}}^{(\mathrm{n})}(\hat{\rho}))_{\mathbf{r}} < 0$, for some $\mathbf{r} \equiv (r_1, \ldots, r_N)$, with $1 \leqslant r_1 < r_2 < \ldots < r_N$.

According to Vogel [24], this criterion (and the following Criterion 3) can also be applied to describe the nonclassicality of space-time correlations and the dynamics of radiation sources by applying the generalized P function:

$$P(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*) = \left\langle \circ\!\circ \prod_{i=1}^{M} \delta(\hat{a}_i - \alpha_i) \circ\!\circ \right\rangle. \tag{9}$$

where $\boldsymbol{\alpha} = (\alpha_1, \ldots, \alpha_M)$, with $\alpha_i = \alpha_i(\mathbf{r}_i, t_i)$ depending on the space-time arguments $(\mathbf{r}_i, t_i)$. By contrast to the standard definition of P function, symbol $\circ\!\circ$ describes both the normal order of field operators and also time order, i.e., time arguments increase to the right (left) in products of creation (annihilation) operators [4]. As an example, we will apply this generalized criterion to show the nonclassicality of photon antibunching and hyperbunching effects in Appendix C.

Note that Criterion 2, even for the choice of $\hat{f}_i$ given by Eq. (2) and in the special case of single-mode fields, does not exactly reduce to the SRV criterion as it appeared in Ref. [22]. To show this, let us denote by $M_N^{(\mathrm{n})}(\hat{\rho})$ the submatrix corresponding to the first N rows and columns of $M^{(\mathrm{n})}(\hat{\rho})$. According to the original SRV criterion (Theorem 3 in Ref. [22]), a single-mode state is nonclassical if there exists an N, such that the leading principal minor is negative, i.e., $\det[M_N^{(\mathrm{n})}(\hat{\rho})] < 0$. Such formulated criterion fails for singular (i.e., $\det[M_N^{(\mathrm{n})}(\hat{\rho})] = 0$) matrices of moments, as explained in detail in the context of quantum entanglement in Ref. [60].

Considering $[M_{\hat{F}}^{(\mathrm{n})}(\hat{\rho})]_{\mathbf{r}}$ is equivalent to considering the correlation matrix corresponding to a subset $\hat{F}' \subset \hat{F}$, with $\hat{F}' = (\hat{f}_{r_1}, \hat{f}_{r_2}, \ldots, \hat{f}_{r_N})$, i.e., $[M_{\hat{F}}^{(\mathrm{n})}(\hat{\rho})]_{\mathbf{r}} = M_{\hat{F}'}^{(\mathrm{n})}(\hat{\rho})$. We note that the subset symbol is used for brevity although it is not very precise, as the $\hat{F}$s are ordered collections of operators.

Thus, by denoting

$$M_{\hat{F}'}^{(\mathrm{n})}(\hat{\rho}) \equiv \left[M_{\hat{F}}^{(\mathrm{n})}(\hat{\rho})\right]_{\mathbf{r}}$$
$$= \begin{pmatrix} \langle :\hat{f}_{r_1}^\dagger \hat{f}_{r_1}: \rangle & \langle :\hat{f}_{r_1}^\dagger \hat{f}_{r_2}: \rangle & \cdots & \langle :\hat{f}_{r_1}^\dagger \hat{f}_{r_N}: \rangle \\ \langle :\hat{f}_{r_2}^\dagger \hat{f}_{r_1}: \rangle & \langle :\hat{f}_{r_2}^\dagger \hat{f}_{r_2}: \rangle & \cdots & \langle :\hat{f}_{r_2}^\dagger \hat{f}_{r_N}: \rangle \\ \vdots & \vdots & \ddots & \vdots \\ \langle :\hat{f}_{r_N}^\dagger \hat{f}_{r_1}: \rangle & \langle :\hat{f}_{r_N}^\dagger \hat{f}_{r_2}: \rangle & \cdots & \langle :\hat{f}_{r_N}^\dagger \hat{f}_{r_N}: \rangle \end{pmatrix}, \tag{10}$$

and its determinant

$$d_{\hat{F}'}^{(\mathrm{n})}(\hat{\rho}) \equiv \det M_{\hat{F}'}^{(\mathrm{n})}(\hat{\rho}), \tag{11}$$

we can equivalently rewrite Criterion 2 as:

Criterion 3. A multimode bosonic state $\hat{\rho}$ is nonclassical if there exists $\hat{F}$ such that $d_{\hat{F}}^{(\mathrm{n})}(\hat{\rho})$ is negative.

This can be written more compactly as:

$$\begin{aligned} \hat{\rho} \text{ is classical } &\Rightarrow \forall \hat{F}: \quad d_{\hat{F}}^{(\mathrm{n})}(\hat{\rho}) \geqslant 0, \\ \hat{\rho} \text{ is nonclassical } &\Leftarrow \exists \hat{F}: \quad d_{\hat{F}}^{(\mathrm{n})}(\hat{\rho}) < 0. \end{aligned} \tag{12}$$

In the following, we use the symbol $\overset{\mathrm{ncl}}{<}$ to emphasize that a given inequality *can* be satisfied only for *nonclassical* states and the symbol $\overset{\mathrm{cl}}{\geqslant}$ to indicate that an inequality *must* be satisfied for all *classical* states.

Let us comment further on the relation between Criteria 2 and 3 and the SRV criterion (in its amended version that takes into account the issue of singular matrices). Criterion 3 corresponds to checking the positivity of an infinite matrix $M_{ij}^{(n)}$ defined as in (6) with the $\hat{f}_i$'s chosen to be all possible monomials given by Eq. (2). Considering the positivity of larger and larger submatrices of this matrix leads to a hierarchy of criteria: testing the positivity of some submatrix $M_N^{(n)}$ leads to a stronger criterion than testing the positivity of a submatrix $M_{N'}^{(n)}$, with $N' < N$. Nonetheless, when one invokes Sylvester's criterion in order to transform the test of positivity of a matrix into the test of positivity of its many principal minors, it is arguably difficult to speak of a "hierarchy." Indeed, because of the issue of the possible singularity of the matrix we cannot simply consider, e.g., leading principal minors involving larger and larger submatrices.

As regards the general formalism, of course by adding operators to the set $\hat{F}$, and therefore increasing the dimension of the matrix $M_{\hat{F}'}^{(n)}$, one obtains a hierarchy of *matrix* conditions on classicality. Nonetheless, also in our case when moving to scalar inequalities by considering determinants, we face the issue of the possible singularity of matrices. Motivated also by this difficulty, in the present article we do not focus so much on the idea a hierarchy of criteria but rather explore the approach that by using matrices of expectations values it is possible to easily obtain criteria of nonclassicality and entanglement in the form of inequalities. As already explained, this is done by referring to Observation 1 and considering $\hat{f}_i$'s possibly more general than monomials, e.g., polynomials.

Indeed, when we choose a set of operators $\hat{F} = (\hat{f}_1, \hat{f}_2, \ldots)$, we compute the corresponding matrix of expectation values, and we check its positivity, what we are doing is equivalent to checking positivity of, e.g., $\langle : \hat{f}^\dagger \hat{f} : \rangle$ for all f's that can be written as a linear combination of the operators in $\hat{F}$: $\hat{f} = \sum_i c_i \hat{f}_i$. As polynomials can be expanded into monomials, it is clear that checking the positivity of a matrix $M^{(n)}_{\hat{F}}$ with $\hat{F}$ consisting of polynomials, cannot give a stronger criterion than checking the positivity of a matrix $M^{(n)}_{\hat{F}'}$, where $\hat{F}'$ is given by all the monomials appearing in the elements of $\hat{F}$. Of course, to have a stronger *matrix* criterion of classicality we pay a price in terms of the dimension of the matrix $M^{(n)}_{\hat{F}'}$, which is larger than $M^{(n)}_{\hat{F}}$. Further, as we will see, by considering general sets $\hat{F}$—that is, not only containing monomials—one can straightforwardly obtain interesting and physically relevant inequalities, which may be difficult to pinpoint when considering monomials as "building blocks." It is worth noting that the possibility of using polynomial functions of moments was also discussed in Ref. [59] in the context of entanglement criterion.

Finally, we remark that to make the above criteria sensitive in detecting nonclassicality, the f_i must be chosen such that the normal ordering is important in giving $M^{(n)}$. In particular, assuming this special structure for the f_i's, there must be some combination of creation and annihilation operators. On the contrary, the inclusion of only creation or only annihilation operators would give a matrix $M^{(n)}$ positive for every state, thus completely useless for detecting nonclassicality.

A. Nonclassicality and the Cauchy-Schwarz inequality

The Cauchy-Schwarz inequality (CSI) for operators can be written as follows (see, e.g., Ref. [5]):

$$\langle \hat{A}^\dagger \hat{A} \rangle \langle \hat{B}^\dagger \hat{B} \rangle \geqslant |\langle \hat{A}^\dagger \hat{B} \rangle|^2, \tag{13}$$

where $\hat{A}$ and $\hat{B}$ are arbitrary operators for which the above expectations exist. Indeed, $\langle \hat{A}^\dagger \hat{B} \rangle \equiv \mathrm{Tr}(\rho \hat{A}^\dagger \hat{B})$ is a valid inner product because of the positivity of ρ. Similarly, one can define a valid scalar product for a positive P function. In detail, by identifying $\hat{A} = f_1(\mathbf{a}, \mathbf{a}^\dagger)$ and $\hat{B} = f_2(\mathbf{a}, \mathbf{a}^\dagger)$, one can define the scalar product

$$\langle : \hat{f}_i^\dagger \hat{f}_j : \rangle = \int d^2\boldsymbol{\alpha}\; f_i^*(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*) f_j(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*) P(\boldsymbol{\alpha}, \boldsymbol{\alpha}^*). \tag{14}$$

Then, a CSI can be written as:

$$\langle : \hat{f}_1^\dagger \hat{f}_1 : \rangle \langle : \hat{f}_2^\dagger \hat{f}_2 : \rangle \overset{\text{cl}}{\geqslant} |\langle : \hat{f}_1^\dagger \hat{f}_2 : \rangle|^2. \tag{15}$$

Such CSI, for a given choice of operators $\hat{f}_1$ and $\hat{f}_2$, can be violated by some nonclassical fields described by a P function which is not positive everywhere, that is, such that (14) does not actually define a scalar product. We then say that the state of the fields violates the CSI. The nonclassicality of states violating the CSI can be shown by analyzing Criterion 3 for $\hat{F} = (\hat{f}_1, \hat{f}_2)$, which results in

$$d^{(\mathrm{n})}_{\hat{F}} = \begin{vmatrix} \langle : \hat{f}_1^\dagger \hat{f}_1 : \rangle & \langle : \hat{f}_1^\dagger \hat{f}_2 : \rangle \\ \langle : \hat{f}_1 \hat{f}_2^\dagger : \rangle & \langle : \hat{f}_2^\dagger \hat{f}_2 : \rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \tag{16}$$

B. A zoo of nonclassical phenomena

In Table I, we present a variety of multimode nonclassicality criteria, which can be derived by applying Criterion 3 as shown in this subsection and in greater detail in Appendices A–C.

In the following, we give a few simple examples of other classical inequalities, which—to our knowledge—have not been discussed in the literature. In particular, we analyze inequalities based on determinants of the following form:

$$D(x,y,z) = \begin{vmatrix} 1 & x & x^* \\ x^* & z & y^* \\ x & y & z \end{vmatrix}. \tag{17}$$

(i) By applying Criterion 3 for $\hat{F} = (1, \hat{a}_1\hat{a}_2, \hat{a}_1^\dagger \hat{a}_2^\dagger)$, we obtain

$$d^{(\mathrm{n})}_{\hat{F}} = D(\langle \hat{a}_1 \hat{a}_2 \rangle, \langle \hat{a}_1^2 \hat{a}_2^2 \rangle, \langle \hat{n}_1 \hat{n}_2 \rangle) \overset{\text{ncl}}{<} 0, \tag{18}$$

where $\hat{n}_1 = \hat{a}_1^\dagger \hat{a}_1$ and $\hat{n}_2 = \hat{a}_2^\dagger \hat{a}_2$. (ii) For $\hat{F} = (1, \hat{a}_1 \hat{a}_2^\dagger, \hat{a}_1^\dagger \hat{a}_2)$ one obtains

$$d^{(\mathrm{n})}_{\hat{F}} = D(\langle \hat{a}_1 \hat{a}_2^\dagger \rangle, \langle \hat{a}_1^2 (\hat{a}_2^\dagger)^2 \rangle, \langle \hat{n}_1 \hat{n}_2 \rangle) \overset{\text{ncl}}{<} 0. \tag{19}$$

(iii) For $\hat{F} = (1, \hat{a}_1 + \hat{a}_2^\dagger, \hat{a}_1^\dagger + \hat{a}_2)$, Criterion 3 leads to

$$d^{(\mathrm{n})}_{\hat{F}} = D(\langle \hat{a}_1 + \hat{a}_2^\dagger \rangle, \langle (\hat{a}_1 + \hat{a}_2^\dagger)^2 \rangle, z) \overset{\text{ncl}}{<} 0, \tag{20}$$

where $z = \langle \hat{n}_1 \rangle + \langle \hat{n}_2 \rangle + 2\mathrm{Re}\langle \hat{a}_1 \hat{a}_2 \rangle$.
(iv) For $\hat{F} = (1, \hat{a}_1 + \hat{a}_2, \hat{a}_1^\dagger + \hat{a}_2^\dagger)$ one has

$$d^{(\mathrm{n})}_{\hat{F}} = D(\langle \hat{a}_1 + \hat{a}_2 \rangle, \langle (\hat{a}_1 + \hat{a}_2)^2 \rangle, z) \overset{\text{ncl}}{<} 0, \tag{21}$$

where $z = \langle \hat{n}_1 \rangle + \langle \hat{n}_2 \rangle + 2\mathrm{Re}\langle \hat{a}_1 \hat{a}_2^\dagger \rangle$. These nonclassicality criteria, given by Eqs. (18)–(21), will be related to the entanglement criteria in subsection III D2.

Another example, which is closely related to the Simon entanglement criterion [68], as will be shown in subsection III D1, can be obtained from Criterion 3 assuming $\hat{F} = (1, \hat{a}_1, \hat{a}_1^\dagger, \hat{a}_2^\dagger, \hat{a}_2)$. Thus, we obtain:

$$d^{(\mathrm{n})}_{\hat{F}} = \begin{vmatrix} 1 & \langle \hat{a}_1 \rangle & \langle \hat{a}_1^\dagger \rangle & \langle \hat{a}_2^\dagger \rangle & \langle \hat{a}_2 \rangle \\ \langle \hat{a}_1^\dagger \rangle & \langle \hat{a}_1^\dagger \hat{a}_1 \rangle & \langle (\hat{a}_1^\dagger)^2 \rangle & \langle \hat{a}_1^\dagger \hat{a}_2^\dagger \rangle & \langle \hat{a}_1^\dagger \hat{a}_2 \rangle \\ \langle \hat{a}_1 \rangle & \langle \hat{a}_1^2 \rangle & \langle \hat{a}_1^\dagger \hat{a}_1 \rangle & \langle \hat{a}_1 \hat{a}_2^\dagger \rangle & \langle \hat{a}_1 \hat{a}_2 \rangle \\ \langle \hat{a}_2 \rangle & \langle \hat{a}_1 \hat{a}_2 \rangle & \langle \hat{a}_1^\dagger \hat{a}_2 \rangle & \langle \hat{a}_2^\dagger \hat{a}_2 \rangle & \langle \hat{a}_2^2 \rangle \\ \langle \hat{a}_2^\dagger \rangle & \langle \hat{a}_1 \hat{a}_2^\dagger \rangle & \langle \hat{a}_1^\dagger \hat{a}_2^\dagger \rangle & \langle (\hat{a}_2^\dagger)^2 \rangle & \langle \hat{a}_2^\dagger \hat{a}_2 \rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \tag{22}$$

III. ENTANGLEMENT AND NONCLASSICALITY CRITERIA

Here, we express various two- and three-mode entanglement inequalities in terms of nonclassicality inequalities derived from Criterion 3, which are summarized in Table II. First, we briefly describe the Shchukin-Vogel entanglement criterion, which enables the derivation of various entanglement inequalities.

A. The Shchukin-Vogel entanglement criterion

The Criterion 3 of nonclassicality resembles the Shchukin-Vogel (SV) criterion [59–61] for distinguishing states with positive partial transposition (PPT) from those with nonpositive partial transposition (NPT). Analogously to Eqs. (7) and (8), one can define a matrix $M(\hat\rho)=[M_{ij}(\hat\rho)]$ of moments as follows:

$$M_{ij}(\hat\rho)=\mathrm{Tr}[(\hat a^{\dagger i_1}\hat a^{i_2}\hat b^{\dagger i_3}\hat b^{i_4})^\dagger(\hat a^{\dagger j_1}\hat a^{j_2}\hat b^{\dagger j_3}\hat b^{j_4})\hat\rho], \tag{23}$$

where the subscripts i and j correspond to multi-indices (i_1,i_2,i_3,i_4) and (j_1,j_2,j_3,j_4), respectively. Note that, contrary to Eq. (7), the creation and annihilation operators are *not* normally ordered. As discussed in Ref. [61], the matrix $M(\hat\rho)$ of moments for a separable state $\hat\rho$ is also separable, i.e.,

$$\hat\rho=\sum_i p_i\hat\rho_i^A\otimes\hat\rho_i^B \Rightarrow M(\hat\rho)=\sum_i p_i M^A\left(\hat\rho_i^A\right)\otimes M^B\left(\hat\rho_i^A\right), \tag{24}$$

where $p_i\geqslant 0$, $\sum_i p_i=1$, $M^A(\hat\rho^A)=\sum_{i'j'}M_{i'j'}(\hat\rho^A)|i'\rangle\langle j'|$ is expressed in a formal basis $\{|i'\rangle\}$ with $i'=(i_1,i_2,0,0)$ and $j'=(j_1,j_2,0,0)$; $M^B(\hat\rho^B)$ defined analogously. Reference [59] proved the following criterion:

Criterion 4. A bipartite quantum state $\hat\rho$ is NPT if and only if $M(\hat\rho^\Gamma)$ is NPT.

The elements of the matrix of moments, $M(\hat\rho^\Gamma)=[M_{ij}(\hat\rho^\Gamma)]$, where Γ denotes partial transposition in some fixed basis, can be simply calculated as

$$\begin{aligned}M_{ij}(\hat\rho^\Gamma)&=\mathrm{Tr}[(\hat a^{\dagger i_1}\hat a^{i_2}\hat b^{\dagger i_3}\hat b^{i_4})^\dagger(\hat a^{\dagger j_1}\hat a^{j_2}\hat b^{\dagger j_3}\hat b^{j_4})\hat\rho^\Gamma]\\&=\mathrm{Tr}[(\hat a^{\dagger i_1}\hat a^{i_2}\hat b^{\dagger j_3}\hat b^{j_4})^\dagger(\hat a^{\dagger j_1}\hat a^{j_2}\hat b^{\dagger i_3}\hat b^{i_4})\hat\rho].\end{aligned} \tag{25}$$

Let us define

$$d_{\hat F}^\Gamma(\hat\rho)=\begin{vmatrix}\langle\hat f_{r_1}^\dagger\hat f_{r_1}\rangle^\Gamma & \langle\hat f_{r_1}^\dagger\hat f_{r_2}\rangle^\Gamma & \cdots & \langle\hat f_{r_1}^\dagger\hat f_{r_N}\rangle^\Gamma\\ \langle\hat f_{r_2}^\dagger\hat f_{r_1}\rangle^\Gamma & \langle\hat f_{r_2}^\dagger\hat f_{r_2}\rangle^\Gamma & \cdots & \langle\hat f_{r_2}^\dagger\hat f_{r_N}\rangle^\Gamma\\ \vdots & \vdots & \ddots & \vdots\\ \langle\hat f_{r_N}^\dagger\hat f_{r_1}\rangle^\Gamma & \langle\hat f_{r_N}^\dagger\hat f_{r_2}\rangle^\Gamma & \cdots & \langle\hat f_{r_N}^\dagger\hat f_{r_N}\rangle^\Gamma\end{vmatrix}, \tag{26}$$

in terms of $\langle\hat f_{r_i}^\dagger\hat f_{r_j}\rangle^\Gamma\equiv\langle(\hat f_{r_i}^\dagger\hat f_{r_j})^\Gamma\rangle$ $(i,j=1,\ldots,N)$. For example, if $\hat X$ is an operator acting on two or more modes, and we take partial transposition with respect to the first mode,

$$\hat X^\Gamma=(T\otimes\mathrm{id})(\hat X),$$

with T the transposition acting on the first mode and id the identity operation doing nothing on the remaining modes, respectively. Then the SV Criterion 4, for brevity referred here to as the entanglement criterion, can be formulated as follows [61]:

Criterion 5. A bipartite state $\hat\rho$ is NPT if and only if there exists $\hat F$, such that $d_{\hat F}^\Gamma(\hat\rho)$ is negative.

This Criterion 5 can be written more compactly as follows:

$$\begin{aligned}\hat\rho \text{ is PPT} &\Leftrightarrow \forall\hat F: \quad d_{\hat F}^\Gamma(\hat\rho)\geqslant 0,\\ \hat\rho \text{ is NPT} &\Leftrightarrow \exists\,\hat F: \quad d_{\hat F}^\Gamma(\hat\rho)<0.\end{aligned} \tag{27}$$

As for the case of the nonclassicality criteria, the original SV criterion actually refers to a set $\hat F$ given by monomials in the creation and annihilation operators. This entanglement criterion can be applied not only to two-mode fields but also to multimode fields [61,76]. Note that Criterion 5 does not detect PPT-entangled states (which are part, and possibly the only members, of the family of the so-called bound entangled states) [77]. Analogously to the notation of $\overset{\text{ncl}}{<}$, we use the symbol $\overset{\text{ent}}{<}$ to indicate that a given inequality can be fulfilled *only* for entangled states.

Here we show that various well-known entanglement inequalities can be derived from the nonclassicality Criterion 3 including the criteria of Hillery and Zubairy [67], Duan *et al.* [66], Simon [68], or Mancini *et al.* [69]. We also derive new entanglement criteria and show their relation to the nonclassicality criterion. Other examples of entanglement inequalities, which can be easily derived from nonclassicality criteria, include [78–80]. However, for brevity, we do not include them here.

B. Entanglement and the Cauchy-Schwarz inequality

The matrix $M_{\hat F}^{(n)}(\hat\rho)$ is linear in its state $\hat\rho=\sum_i p_i\hat\rho_i$. Therefore we have

$$M_{\hat F}^{(n)}(\hat\rho)=\sum_i p_i M_{\hat F}^{(n)}(\hat\rho_i)\geqslant 0 \tag{28}$$

if $M_{\hat F}^{(n)}(\hat\rho_i)\geqslant 0$ for all $\hat\rho_i$. Thus, $M_{\hat F}^{(n)}$ is positive for separable states if it is positive on factorized states.

Let

$$\hat F=(\hat f_1,\ldots,\hat f_N) \tag{29}$$

with functions $\hat f_i=\hat f_{i1}\hat f_{i2}\cdots\hat f_{iM}$, where

$$\hat f_{ij}=\begin{cases}1 & \text{if } i\neq k_j\\ \text{either } g_j(\hat a_j)\text{ or } g_j(\hat a_j^\dagger) & \text{if } i=k_j.\end{cases} \tag{30}$$

Here, i is the index of the element $\hat f_i$ in $\hat F$, and index j refers to the mode. $\hat f_{ij}$ is possibly different from the identity for one unique value $i=k_j$, and in that case it is equal to a function g_j of either the creation or annihilation operators of mode j, but not of both.

Writing the matrix $M_{\hat F}^{(n)}$ in a formal basis $\{|k\rangle\}$, one then has

$$\begin{aligned}M_{\hat F}^{(n)}&=\sum_{kl}\langle:\hat f_k^\dagger\hat f_l:\rangle|k\rangle\langle l|\\&=\sum_{kl}\langle:\hat f_{k1}^\dagger\hat f_{l1}\ldots\hat f_{kM}^\dagger\hat f_{lM}:\rangle|k\rangle\langle l|.\end{aligned} \tag{31}$$

For factorized states holds

$$\begin{aligned}M_{\hat F}^{(n)}&=\sum_{kl}\langle:\hat f_{k1}^\dagger\hat f_{l1}:\rangle\cdots\langle:\hat f_{kM}^\dagger\hat f_{lM}:\rangle|k\rangle\langle l|\\&=\sum_{k}\langle:\hat f_{k1}^\dagger\hat f_{k1}:\rangle\cdots\langle:\hat f_{kM}^\dagger\hat f_{kM}:\rangle|k\rangle\langle l|\\&\quad+\sum_{k\neq l}\langle:\hat f_{k1}^\dagger\hat f_{l1}:\rangle\cdots\langle:\hat f_{kM}^\dagger\hat f_{lM}:\rangle|k\rangle\langle l|\\&=\sum_{k}\langle:\hat f_{k1}^\dagger\hat f_{k1}:\rangle\cdots\langle:\hat f_{kM}^\dagger\hat f_{kM}:\rangle|k\rangle\langle l|\end{aligned}$$

$$
\begin{aligned}
&+\sum_{k\neq l}\langle \hat{f}_{k1}^{\dagger}\rangle\langle \hat{f}_{l1}\rangle\cdots\langle \hat{f}_{kM}^{\dagger}\rangle\langle \hat{f}_{lM}\rangle|k\rangle\langle l|\\
\geqslant& \sum_{k}\langle \hat{f}_{k1}^{\dagger}\rangle\langle \hat{f}_{k1}\rangle\cdots\langle \hat{f}_{kM}^{\dagger}\rangle\langle \hat{f}_{kM}\rangle|k\rangle\langle l|\\
&+\sum_{k\neq l}\langle \hat{f}_{k1}^{\dagger}\rangle\langle \hat{f}_{l1}\rangle\cdots\langle \hat{f}_{kM}^{\dagger}\rangle\langle \hat{f}_{lM}\rangle|k\rangle\langle l|\\
=&\left(\sum_{k}\langle \hat{f}_{k1}^{\dagger}\rangle\cdots\langle \hat{f}_{kM}^{\dagger}\rangle|k\rangle\right)\\
&\times\left(\sum_{l}\langle \hat{f}_{l1}\rangle\cdots\langle \hat{f}_{lM}\rangle\langle l|\right)\geqslant 0. \qquad (32)
\end{aligned}
$$

The first equality comes from the state being factorized. The third equality is due to the fact that the $\hat{f}_{ij}$s are functions of either annihilation or creation operators, but not of both, so $\langle :\hat{f}_{k1}^{\dagger}\hat{f}_{l1}:\rangle = \langle \hat{f}_{k1}^{\dagger}\hat{f}_{l1}\rangle$ or $\langle :\hat{f}_{k1}^{\dagger}\hat{f}_{l1}:\rangle = \langle \hat{f}_{l1}\hat{f}_{k1}^{\dagger}\rangle$, and that for $k\neq l$ at least one among $\hat{f}_{k1}^{\dagger}$ and $\hat{f}_{l1}$, let us say, e.g., $\hat{f}_{l1}$, is equal to the identity—in particular this implies that its expectation value is equal to $\langle \hat{f}_{l1}\rangle = 1$. The first inequality is due to the fact that $\langle :\hat{f}_{k1}^{\dagger}\hat{f}_{k1}:\rangle = \langle \hat{f}_{k1}^{\dagger}\hat{f}_{k1}\rangle$ or $\langle :\hat{f}_{k1}^{\dagger}\hat{f}_{k1}:\rangle = \langle \hat{f}_{k1}\hat{f}_{k1}^{\dagger}\rangle$ and to the Cauchy-Schwarz inequality.

C. Entanglement criteria *equal* to nonclassicality criteria

By applying the nonclassicality Criterion 3, we give a few examples of classical inequalities, which can be violated *only* by entangled states.

1. Hillery-Zubairy's entanglement criteria

Hillery and Zubairy [67] derived a few entanglement inequalities both for two-mode fields:

$$\langle \hat{n}_1\hat{n}_2\rangle \overset{\text{ent}}{<} |\langle \hat{a}_1\hat{a}_2^{\dagger}\rangle|^2, \qquad (33)$$

$$\langle \hat{n}_1\rangle\langle \hat{n}_2\rangle \overset{\text{ent}}{<} |\langle \hat{a}_1\hat{a}_2\rangle|^2, \qquad (34)$$

and three-mode fields

$$\langle \hat{n}_1\hat{n}_2\hat{n}_3\rangle \overset{\text{ent}}{<} |\langle \hat{a}_1^{\dagger}\hat{a}_2\hat{a}_3\rangle|^2. \qquad (35)$$

These inequalities can be derived from the entanglement Criterion 5 [59,61] assuming: $\hat{F} = (1,\hat{a}_1\hat{a}_2)$ to derive Eq. (33), $\hat{F} = (\hat{a}_1,\hat{a}_2)$ for Eq. (34), and $\hat{F} = (1,\hat{a}_1\hat{a}_2\hat{a}_3)$ for Eq. (35).

On the other hand, Eq. (33) can be obtained from the nonclassicality Criterion 3 assuming $\hat{F} = (1,\hat{a}_1\hat{a}_2^{\dagger})$, which gives

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} 1 & \langle \hat{a}_1\hat{a}_2^{\dagger}\rangle \\ \langle \hat{a}_1^{\dagger}\hat{a}_2\rangle & \langle \hat{n}_1\hat{n}_2\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \qquad (36)$$

Analogously, assuming $\hat{F} = (\hat{a}_1,\hat{a}_2^{\dagger})$, one gets

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} \langle \hat{n}_1\rangle & \langle \hat{a}_1^{\dagger}\hat{a}_2^{\dagger}\rangle \\ \langle \hat{a}_1\hat{a}_2\rangle & \langle \hat{n}_2\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0, \qquad (37)$$

which corresponds to Eq. (34). By choosing a set of three-mode operators $\hat{F} = (1,\hat{a}_1^{\dagger}\hat{a}_2\hat{a}_3)$, one readily obtains

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} 1 & \langle \hat{a}_1^{\dagger}\hat{a}_2\hat{a}_3\rangle \\ \langle \hat{a}_1\hat{a}_2^{\dagger}\hat{a}_3^{\dagger}\rangle & \langle \hat{n}_1\hat{n}_2\hat{n}_3\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0, \qquad (38)$$

which corresponds to Eq. (35).

By applying Criterion 3 with $\hat{F} = (\hat{a}_1^{\dagger},\hat{a}_2\hat{a}_3)$, we find another inequality

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} \langle \hat{n}_1\rangle & \langle \hat{a}_1\hat{a}_2\hat{a}_3\rangle \\ \langle \hat{a}_1\hat{a}_2\hat{a}_3\rangle^* & \langle \hat{n}_2\hat{n}_3\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0, \qquad (39)$$

which was derived in Ref. [61] from the entanglement Criterion 5.

Using the Cauchy-Schwarz inequality, Hillery and Zubairy [67] also found a more general form of inequality than the one in Eq. (33), which reads as follows:

$$\left\langle (\hat{a}_1^{\dagger})^m\hat{a}_1^m(\hat{a}_2^{\dagger})^n\hat{a}_2^n\right\rangle \overset{\text{ent}}{<} \left|\left\langle \hat{a}_1^m(\hat{a}_2^{\dagger})^n\right\rangle\right|^2. \qquad (40)$$

This inequality can be derived from the nonclassicality Criterion 3 for $\hat{F} = \mathbf{(}1,\hat{a}_1^m(\hat{a}_2^{\dagger})^n\mathbf{)}$, which leads to

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} 1 & \left\langle \hat{a}_1^m(\hat{a}_2^{\dagger})^n\right\rangle \\ \left\langle (\hat{a}_1^{\dagger})^m\hat{a}_2^n\right\rangle & \left\langle (\hat{a}_1^{\dagger})^m\hat{a}_1^m(\hat{a}_2^{\dagger})^n\hat{a}_2^n\right\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \qquad (41)$$

Alternatively, Eq. (40) can be derived from the entanglement Criterion 5 for $\hat{F} = (1,\hat{a}_1^m\hat{a}_2^n)$. Thus, we see that

$$d^{(\text{n})}\left[1,\hat{a}_1^m(\hat{a}_2^{\dagger})^n\right] = d^{\Gamma}(1,\hat{a}_1^m\hat{a}_2^n) \overset{\text{ent}}{<} 0, \qquad (42)$$

where, for clarity, we use the notation $d^k(\hat{F})$ instead of $d_{\hat{F}}^k$ for $k = (n),\Gamma$. Moreover, we can generalize entanglement inequality, given by Eq. (38), as follows:

$$\left\langle \hat{n}_1^k\hat{n}_2^l\hat{n}_3^m\right\rangle \overset{\text{ent}}{<} \left|\left\langle (\hat{a}_1^{\dagger})^k\hat{a}_2^l\hat{a}_3^m\right\rangle\right|^2 \qquad (43)$$

for arbitrary integers $k,l,m > 0$. This inequality can be proved by applying both Criteria 3 and 5:

$$
\begin{aligned}
d^{(\text{n})}\left[1,(\hat{a}_1^{\dagger})^k\hat{a}_2^l\hat{a}_3^m\right] &= d^{\Gamma}\left(1,\hat{a}_1^k\hat{a}_2^l\hat{a}_3^m\right)\\
&= \begin{vmatrix} 1 & \left\langle (\hat{a}_1^{\dagger})^k\hat{a}_2^l\hat{a}_3^m\right\rangle \\ \left\langle (\hat{a}_1^{\dagger})^k\hat{a}_2^l\hat{a}_3^m\right\rangle^* & \left\langle \hat{n}_1^k\hat{n}_2^l\hat{n}_3^m\right\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0, \qquad (44)
\end{aligned}
$$

where the first mode is partially transposed. Analogously, Eq. (39) can be generalized to following entanglement inequality:

$$\left\langle \hat{n}_1^k\right\rangle\left\langle \hat{n}_2^l\hat{n}_3^m\right\rangle \overset{\text{ent}}{<} \left|\left\langle \hat{a}_1^k\hat{a}_2^l\hat{a}_3^m\right\rangle\right|^2, \qquad (45)$$

which can be shown by applying Criteria 3 and 5:

$$
\begin{aligned}
d^{(\text{n})}\left[(\hat{a}_1^{\dagger})^k,\hat{a}_2^l\hat{a}_3^m\right] &= d^{\Gamma}\left(\hat{a}_1^k,\hat{a}_2^l\hat{a}_3^m\right)\\
&= \begin{vmatrix} \left\langle \hat{n}_1^k\right\rangle & \left\langle \hat{a}_1^k\hat{a}_2^l\hat{a}_3^m\right\rangle \\ \left\langle \hat{a}_1^k\hat{a}_2^l\hat{a}_3^m\right\rangle^* & \left\langle \hat{n}_2^l\hat{n}_3^m\right\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \qquad (46)
\end{aligned}
$$

It is worth remarking that in all the above cases, once the $\overset{\text{ncl}}{<}$ inequalities are found as nonclassicality inequalities, it is easy to check that they can be satisfied only by entangled states; that is, they really are $\overset{\text{ent}}{<}$ inequalities. Indeed, the determinant condition is the only nontrivial one for establishing the positivity of the involved 2×2 matrices. Further, these matrices are linear in the state with respect to which the expectation values are calculated. Thus, if we prove that the matrices are positive for factorized states, then we have that they are necessarily positive for a separable state and so are the determinants. For the sake of concreteness and clarity, we prove the positivity of the 2×2 matrix of Eq. (37) for a factorized state. The positivity of the other matrices for factorized states is analogously proved.

For a factorized state, as a special case of inequalities given in Eq. (32), we have

$$\begin{aligned}\begin{pmatrix} \langle\hat{n}_1\rangle & \langle\hat{a}_1^\dagger\hat{a}_2^\dagger\rangle \\ \langle\hat{a}_1\hat{a}_2\rangle & \langle\hat{n}_2\rangle \end{pmatrix} &= \begin{pmatrix} \langle\hat{a}_1^\dagger\hat{a}_1\rangle & \langle\hat{a}_1^\dagger\rangle\langle\hat{a}_2^\dagger\rangle \\ \langle\hat{a}_1\rangle\langle\hat{a}_2\rangle & \langle\hat{a}_2^\dagger\hat{a}_2\rangle \end{pmatrix} \\ &\geqslant \begin{pmatrix} \langle\hat{a}_1^\dagger\rangle\langle\hat{a}_1\rangle & \langle\hat{a}_1^\dagger\rangle\langle\hat{a}_2^\dagger\rangle \\ \langle\hat{a}_1\rangle\langle\hat{a}_2\rangle & \langle\hat{a}_2^\dagger\rangle\langle\hat{a}_2\rangle \end{pmatrix} \\ &= \begin{pmatrix} \langle\hat{a}_1^\dagger\rangle \\ \langle\hat{a}_2\rangle \end{pmatrix} (\langle\hat{a}_1\rangle \quad \langle\hat{a}_2^\dagger\rangle) \geqslant 0, \end{aligned} \tag{47}$$

where the first inequality is due to the Cauchy-Schwarz inequality $\langle\hat{X}^\dagger\hat{X}\rangle \geqslant |\langle\hat{X}\rangle|^2$.

2. *Entanglement criterion of Duan et al.*

A sharpened version of the entanglement criterion of Duan *et al.* [66] can be formulated as follows [59]:

$$\langle\Delta\hat{a}_1^\dagger\Delta\hat{a}_1\rangle\langle\Delta\hat{a}_2^\dagger\Delta\hat{a}_2\rangle \overset{\text{ent}}{<} |\langle\Delta\hat{a}_1\Delta\hat{a}_2\rangle|^2, \tag{48}$$

where $\Delta\hat{a}_i = \hat{a}_i - \langle\hat{a}_i\rangle$ for $i = 1,2$. Equation (48) follows from the entanglement Criterion 5 for $\hat{F} = (1,\hat{a}_1,\hat{a}_2)$ [59] or, equivalently, for $\hat{F} = (\Delta\hat{a}_1,\Delta\hat{a}_2)$. It can also be derived from the nonclassicality Criterion 3 for $\hat{F} = (\Delta\hat{a}_1,\Delta\hat{a}_2^\dagger)$. Thus, we obtain

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} \langle\Delta\hat{a}_1^\dagger\Delta\hat{a}_1\rangle & \langle\Delta\hat{a}_1^\dagger\Delta\hat{a}_2^\dagger\rangle \\ \langle\Delta\hat{a}_1\Delta\hat{a}_2\rangle & \langle\Delta\hat{a}_2^\dagger\Delta\hat{a}_2\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0, \tag{49}$$

which corresponds to Eq. (48). Alternatively, by choosing $\hat{F} = (1,\hat{a}_1,\hat{a}_2^\dagger)$, one obtains

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} 1 & \langle\hat{a}_1\rangle & \langle\hat{a}_2^\dagger\rangle \\ \langle\hat{a}_1^\dagger\rangle & \langle\hat{n}_1\rangle & \langle\hat{a}_1^\dagger\hat{a}_2^\dagger\rangle \\ \langle\hat{a}_2\rangle & \langle\hat{a}_1\hat{a}_2\rangle & \langle\hat{n}_2\rangle \end{vmatrix}, \tag{50}$$

which is equal to Eq. (49). Thus, it is seen that this nonclassicality criterion is equal to the entanglement criterion. Moreover, the advantage of using polynomials, instead of monomial, functions of moments in $\hat{F}$ is apparent. The same conclusion was drawn by comparing Eqs. (A9) and (A10) or Eqs. (C12) and (C13).

D. Entanglement criteria via sums of nonclassicality criteria

Here, we present a few examples of classical inequalities derived from the entanglement Criterion 5 and the nonclassicality Criterion 3 that are apparently not equal. More specifically, we have presented in subsection III C examples of classical inequalities, which can be derived from the entanglement Criterion 5 for a given $\hat{F}_1$ or, equivalently, from the nonclassicality Criterion 3 for $\hat{F}_2$ equal to a partial transpose of $\hat{F}_1$. In this section, we give examples of entanglement inequalities, which *cannot* be derived from Criterion 3 for $\hat{F}_2 = \hat{F}_1^\Gamma$.

States satisfying Criterion 5 for entanglement must be nonclassical, as any entangled state is necessary nonclassical in the sense of Criterion 1. We will provide specific examples that satisfying an entanglement inequality implies satisfying one or more nonclassical inequalities. This approach enables an analysis of the entanglement for a given nonclassicality. The main problem is to express $d_{\hat{F}}^\Gamma \equiv d^\Gamma(\hat{F})$ as linear combinations of some $d^{(\text{n})}(\hat{F}^{(k)})$, i.e.:

$$d_{\hat{F}}^\Gamma = \sum_k c_k d^{(\text{n})}(\hat{F}^{(k)}), \tag{51}$$

where $c_k > 0$. To find such expansions explicitly, we apply the following three properties of determinants: (i) The Laplace expansion formula along any row (or column): $\det M = \sum_j (-1)^{i+j} M_{ij}\mu_{ij}$, where μ_{ij} is a minor of a matrix $M = (M_{ij})$. (ii) Swapping rule: By exchanging any two rows (columns) of a determinant, the value of the determinant is the same of the original determinant but with opposite sign. (iii) Summation rule: If some (or all) the elements of a column (row) are sum of two terms, then the determinant can be given as the sum of two determinants, e.g., $\det(a+a',b+b';c,d) = \det(a,b;c,d) + \det(a',b';c,d)$.

1. *Simon's entanglement criterion*

As the first example of such nontrivial relation between the nonclassicality and entanglement criteria, let us consider Simon's entanglement criterion [68]. As shown in Ref. [59], it can be obtained from Criterion 5 as $d_{\hat{F}}^\Gamma \overset{\text{ent}}{<} 0$ for $\hat{F} = (1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2,\hat{a}_2^\dagger)$. We found that Simon's criterion can be expressed as a sum of nonclassicality criteria as follows:

$$\begin{aligned} d_{\hat{F}}^\Gamma &= d^{(\text{n})}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger,\hat{a}_2) + d^{(\text{n})}(1,\hat{a}_1,\hat{a}_2^\dagger) \\ &\quad + d^{(\text{n})}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger) + d^{(\text{n})}(1,\hat{a}_1,\hat{a}_2^\dagger,\hat{a}_2), \end{aligned} \tag{52}$$

where $d^{(\text{n})}(1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger,\hat{a}_2)$ is given by Eq. (22). Moreover, $d_{\hat{F}}^{(\text{n})}$ for $\hat{F} = (1,\hat{a}_1,\hat{a}_1^\dagger,\hat{a}_2^\dagger)$, $\hat{F} = (1,\hat{a}_1,\hat{a}_2^\dagger,\hat{a}_2)$, and $\hat{F} = (1,\hat{a}_1,\hat{a}_2^\dagger)$ can be obtained from (22) by analyzing its principal minors. Thus, one can prove the entanglement for a given nonclassicality by checking the violation of specific classical inequalities resulting from the nonclassicality Criterion 3.

2. *Other entanglement criteria*

Now, we present a few entanglement inequalities, which are simpler than Simon's criterion but still correspond to sums of nonclassicality inequalities.

Let us denote the following determinant:

$$D(x,y,z,z') = \begin{vmatrix} 1 & x & x^* \\ x^* & z & y^* \\ x & y & z' \end{vmatrix}. \tag{53}$$

(i) Criterion 5 for $\hat{F} = (1,\hat{a}_1\hat{a}_2,\hat{a}_1^\dagger\hat{a}_2^\dagger)$ results in

$$d_{\hat{F}}^\Gamma = D\left(\langle\hat{a}_1\hat{a}_2^\dagger\rangle,\langle\hat{a}_1^2\hat{a}_2^{\dagger 2}\rangle,\langle\hat{n}_1\hat{n}_2\rangle,z'\right) \overset{\text{ent}}{<} 0, \tag{54}$$

where $z' = \langle(\hat{n}_1+1)(\hat{n}_2+1)\rangle$. By using the aforementioned properties of determinants, we find that the entanglement criterion in Eq. (54) can be given as the following sum of nonclassicality inequalities resulting from Criterion 3:

$$\begin{aligned} d_{\hat{F}}^\Gamma &= d^{(\text{n})}(1,\hat{a}_1\hat{a}_2^\dagger,\hat{a}_1^\dagger\hat{a}_2) \\ &\quad + (\langle\hat{n}_1\rangle + \langle\hat{n}_2\rangle + 1)\, d^{(\text{n})}(1,\hat{a}_1\hat{a}_2^\dagger). \end{aligned} \tag{55}$$

(ii) Criterion 5 for $\hat{F} = (1,\hat{a}_1\hat{a}_2^\dagger,\hat{a}_1^\dagger\hat{a}_2)$ leads to

$$d_{\hat{F}}^\Gamma = D(\langle\hat{a}_1\hat{a}_2\rangle,\langle\hat{a}_1^2\hat{a}_2^2\rangle,z,z') \overset{\text{ent}}{<} 0, \tag{56}$$

where $z = \langle\hat{n}_1\hat{n}_2\rangle + \langle\hat{n}_1\rangle$ and $z' = \langle\hat{n}_1\hat{n}_2\rangle + \langle\hat{n}_2\rangle$. Analogously to Eq. (55), we find that the following sum of the nonclassicality criteria corresponds to the entanglement criterion in Eq. (56):

$$d_{\hat{F}}^{\Gamma} = d^{(n)}(1,\hat{a}_1\hat{a}_2,\hat{a}_1^\dagger\hat{a}_2^\dagger) + \langle\hat{n}_1\rangle\langle\hat{n}_2\rangle + (\langle\hat{n}_1\rangle + \langle\hat{n}_2\rangle)d^{(n)}(1,\hat{a}_1\hat{a}_2). \tag{57}$$

(iii) For $\hat{F} = (1,\hat{a}_1 + \hat{a}_2^\dagger,\hat{a}_1^\dagger + \hat{a}_2)$, one obtains

$$d_{\hat{F}}^{\Gamma} = D(\langle\hat{a}_1 + \hat{a}_2\rangle,\langle(\hat{a}_1 + \hat{a}_2)^2\rangle,z,z) \overset{\text{ent}}{<} 0, \tag{58}$$

where $z = \langle\hat{n}_1\rangle + \langle\hat{n}_2\rangle + 2\text{Re}\langle\hat{a}_1\hat{a}_2^\dagger\rangle + 1$. Analogously to the former cases, we find the relation between the entanglement criterion in Eq. (58) and the nonclassicality Criterion 3 as follows:

$$d_{\hat{F}}^{\Gamma} = d^{(n)}(1,\hat{a}_1 + \hat{a}_2,\hat{a}_1^\dagger + \hat{a}_2^\dagger) + 2d^{(n)}(1,\hat{a}_1 + \hat{a}_2) + 1. \tag{59}$$

(iv) As a final example, let us consider the entanglement Criterion 5 for $\hat{F} = (1,\hat{a}_1 + \hat{a}_2,\hat{a}_1^\dagger + \hat{a}_2^\dagger)$. One obtains

$$d_{\hat{F}}^{\Gamma} = D(\langle\hat{a}_1 + \hat{a}_2^\dagger\rangle,\langle(\hat{a}_1 + \hat{a}_2^\dagger)^2\rangle,z,z') \overset{\text{ent}}{<} 0, \tag{60}$$

where $z = \langle\hat{n}_1\rangle + \langle\hat{n}_2\rangle + 2\text{Re}\langle\hat{a}_1\hat{a}_2\rangle$, and $z' = z + 2$, which is related to the nonclassicality Criterion 3 as follows:

$$d_{\hat{F}}^{\Gamma} = d^{(n)}(1,\hat{a}_1 + \hat{a}_2^\dagger,\hat{a}_1^\dagger + \hat{a}_2) + 2d^{(n)}(1,\hat{a}_1 + \hat{a}_2^\dagger), \tag{61}$$

where $d^{(n)}(1,\hat{a}_1 + \hat{a}_2^\dagger,\hat{a}_1^\dagger + \hat{a}_2)$ is given by Eq. (20), and $d^{(n)}(1,\hat{a}_1 + \hat{a}_2^\dagger)$ is given by its principal minor. Equation (60) corresponds to the entanglement criterion of Mancini *et al.* [69] (see also Ref. [59]).

IV. CONCLUSIONS

We derived classical inequalities for multimode bosonic fields, which can *only* be violated by *nonclassical* fields, so they can serve as a nonclassicality (or quantumness) test. Our criteria are based on Vogel's criterion [24], which is a generalization of analogous criteria for single-mode fields of Agarwal and Tara [20] and, more directly, of Shchukin, Richter, and Vogel (SRV) [21,22]. The nonclassicality criteria correspond to analyzing the positivity of matrices of normally ordered moments of, e.g., annihilation and creation operators, which, by virtue of Sylvester's criterion, correspond to analyzing the positivity of Glauber-Sudarshan *P* function. We used not only monomial but also polynomial functions of moments. We showed that this approach can enable simpler and more intuitive derivation of physically relevant inequalities.

We demonstrated how the nonclassicality criteria introduced here easily reduce to the well-known inequalities (see, e.g., textbooks [3–6], reviews [7–9,11], and Refs. [25–41]) describing various multimode nonclassical effects, for short referred to as the nonclassicality inequalities. Our examples, summarized in Tables I and II, include the following:

(i) Multimode quadrature squeezing [4] and its generalizations, including the sum and difference squeezing defined by Hillery [33], and An and Tinh [39,40], as well the principal squeezing related to the Schrödinger-Robertson indeterminacy relation [81] as defined by Lukš *et al.* [32].

(ii) Single-time photon-number correlations of two modes, including squeezing of the sum and difference of photon numbers (which is also referred to as the photon-number sum/difference sub-Poisson photon-number statistics) [6], violations of the Cauchy-Schwarz inequality [5] and violations of the Muirhead inequality [34,82], which is a generalization of the arithmetic-geometric mean inequality.

(iii) Two-time photon-number correlations of single modes including photon antibunching [4,5,37] and photon hyperbunching [38,41] for stationary and nonstationary fields.

(iv) Two- and three-mode quantum entanglement inequalities (e.g., Refs. [66–69]). We have shown that some known entanglement inequalities (e.g., of Duan *et al.* [66] and Hillery and Zubairy [67]) can be derived as nonclassical inequalities. Other entanglement inequalities (e.g., of Simon [68]) can be represented by sums of nonclassicality inequalities.

Moreover, we developed a general method of expressing inequalities derived from the Shchukin-Vogel entanglement criterion [59,60] as a sum of inequalities derived from the nonclassicality criteria. This approach enables a deeper analysis of the entanglement for a given nonclassicality. We also presented a few inequalities derived from the nonclassicality and entanglement criteria, which to our knowledge have not yet been described in the literature.

It is seen that the nonclassicality criteria based on matrices of moments offer an effective way to derive specific inequalities which might be useful in the verification of nonclassicality of particular states generated in experiments. It seems that the quantum-information community more or less ignores nonclassicality as something closely related to quantum entanglement. We hope that this article presents a useful approach in the direction of a common treatment of both types of phenomena.

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APPENDIX A: UNIFIED DERIVATIONS OF CRITERIA FOR QUADRATURE SQUEEZING AND ITS GENERALIZATIONS

Here and in the following appendices, we present a unified derivation of the known criteria for various multimode nonclassicality phenomena, which are summarized in Table I.

1. Multimode quadrature squeezing

The *quadrature squeezing* of multimode fields can be defined by a negative value of the normally ordered variance [4,9,27]

$$\langle:(\Delta\hat{X}_\phi)^2:\rangle < 0 \quad \text{(A1)}$$

with $\Delta\hat{X}_\phi = \hat{X}_\phi - \langle\hat{X}_\phi\rangle$, of the multimode quadrature operator

$$\hat{X}_\phi = \sum_{m=1}^{M} c_m\, \hat{x}_m(\phi_m), \quad \text{(A2)}$$

which is given in terms of single-mode phase-rotated quadratures given by

$$\hat{x}_m(\phi_m) = \hat{a}_m \exp(i\phi_m) + \hat{a}_m^\dagger \exp(-i\phi_m). \quad \text{(A3)}$$

It is a straightforward generalization of the single-mode quadrature squeezing [7,25]. In (A2), $\boldsymbol{\phi} = (\phi_1,\ldots,\phi_M)$ and c_m are real parameters. In the analysis of physical systems, it is convenient to analyze the annihilation ($\hat{a}_m$) and creation ($\hat{a}_m^\dagger$) operators corresponding to slowly varying operators. Usually, $\hat{x}_m(0)$ and $\hat{x}_m(\pi/2)$ are interpreted as canonical position and momentum operators, although this interpretation can be applied for any two quadratures of orthogonal phases, $\hat{x}_m(\phi_m)$ and $\hat{x}_m(\phi_m+\pi/2)$.

The normally ordered variance can be directly calculated from the P function as follows:

$$\langle:(\Delta\hat{X}_\phi)^2:\rangle = \int d^2\boldsymbol{\alpha}\; P(\boldsymbol{\alpha},\boldsymbol{\alpha}^*)[X_\phi(\boldsymbol{\alpha},\boldsymbol{\alpha}^*) - \langle\hat{X}_\phi\rangle]^2, \quad \text{(A4)}$$

where

$$X_\phi(\boldsymbol{\alpha},\boldsymbol{\alpha}^*) = \sum_{m=1}^{M} c_m(\alpha_m e^{i\phi_m} + \alpha_m^* e^{-i\phi_m}) \quad \text{(A5)}$$

and $\boldsymbol{\alpha} = (\alpha_1,\ldots,\alpha_M)$. From Eq. (A4) it is seen that a negative value of $\langle:(\Delta\hat{X}_\phi)^2:\rangle$ implies the nonpositivity of the P function in some regions of phase space, so the multimode quadrature squeezing is a nonclassical effect. This conclusion can also be drawn by applying Criterion 3. In fact, by choosing $\hat{F} = (1,\hat{X}_\phi)$, one gets

$$d_{\hat{F}}^{(n)} = \begin{vmatrix} 1 & \langle\hat{X}_\phi\rangle \\ \langle\hat{X}_\phi\rangle & \langle:\hat{X}_\phi^2:\rangle \end{vmatrix} = \langle:(\Delta\hat{X}_\phi)^2:\rangle \overset{\text{ncl}}{<} 0, \quad \text{(A6)}$$

which is the squeezing condition (A1).

2. Two-mode principal squeezing

For simplicity, we analyze below the two-mode ($M=2$) case for $c_1=c_2=1$ and $\phi_2-\phi_1=\pi/2$. The two-mode *principal* (quadrature) squeezing can be defined as the $\boldsymbol{\phi}$-optimized squeezing defined by Eq. (A1):

$$\min_{\phi:\phi_2-\phi_1=\pi/2} \langle:(\Delta\hat{X}_\phi)^2:\rangle < 0. \quad \text{(A7)}$$

By applying the Schrödinger-Robertson indeterminacy relation [81], Lukš *et al.* [32] have given the following necessary and sufficient condition for the two-mode *principal squeezing*

$$\langle\Delta\hat{a}_{12}^\dagger\Delta\hat{a}_{12}\rangle < |\langle(\Delta\hat{a}_{12})^2\rangle|, \quad \text{(A8)}$$

where

$$\hat{a}_{12} = \hat{a}_1 + \hat{a}_2, \quad \Delta\hat{a}_{12} = \hat{a}_{12} - \langle\hat{a}_{12}\rangle.$$

This condition for principal squeezing can be derived from Criterion 3 by choosing $\hat{F} = (\Delta\hat{a}_{12}^\dagger,\Delta\hat{a}_{12})$, which leads to:

$$d_{\hat{F}}^{(n)} = \begin{vmatrix} \langle\Delta\hat{a}_{12}^\dagger\Delta\hat{a}_{12}\rangle & \langle(\Delta\hat{a}_{12})^2\rangle \\ \langle(\Delta\hat{a}_{12}^\dagger)^2\rangle & \langle\Delta\hat{a}_{12}^\dagger\Delta\hat{a}_{12}\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \quad \text{(A9)}$$

Equivalently, by applying Criterion 3 for $\hat{F} = (1,\hat{a}_{12}^\dagger,\hat{a}_{12})$ one obtains:

$$d_{\hat{F}}^{(n)} = \begin{vmatrix} 1 & \langle\hat{a}_{12}^\dagger\rangle & \langle\hat{a}_{12}\rangle \\ \langle\hat{a}_{12}\rangle & \langle\hat{n}_{12}\rangle & \langle(\hat{a}_{12})^2\rangle \\ \langle\hat{a}_{12}^\dagger\rangle & \langle(\hat{a}_{12}^\dagger)^2\rangle & \langle\hat{n}_{12}\rangle \end{vmatrix}, \quad \text{(A10)}$$

where

$$\hat{n}_{12} = \hat{a}_{12}^\dagger\hat{a}_{12} = \hat{n}_1 + \hat{n}_2 + 2\mathrm{Re}(\hat{a}_1^\dagger\hat{a}_2).$$

The determinants, given by Eqs. (A9) and (A10) are equal to each other and equivalent to Eq. (A8). This example shows that the application of polynomial functions of moments, instead of monomials, can lead to matrices of moments of lower dimension. Thus, the polynomial-based approach can enable simpler and more intuitive derivations of physically relevant criteria.

3. Sum squeezing

According to Hillery [33], a two-mode state exhibits *sum squeezing* in the direction ϕ if the variance of

$$\hat{V}_\phi = \tfrac{1}{2}(\hat{a}_1\hat{a}_2 e^{-i\phi} + \hat{a}_1^\dagger\hat{a}_2^\dagger e^{i\phi}) \quad \text{(A11)}$$

satisfies

$$\langle(\Delta\hat{V}_\phi)^2\rangle < \tfrac{1}{2}\langle\hat{V}_z\rangle, \quad \text{(A12)}$$

where

$$\hat{V}_z = \tfrac{1}{2}(\hat{n}_1 + \hat{n}_2 + 1)$$

and $\hat{n}_m = \hat{a}_m^\dagger\hat{a}_m$ for $m=1,2$. As for the case of quadrature squeezing, $\hat{a}_1$ and $\hat{a}_2$ usually correspond to slowly varying operators. Let us denote $\hat{V}_x = \hat{V}(\phi=0)$ and $\hat{V}_y = \hat{V}(\phi=\pi/2)$. It is worth mentioning that the operators $\hat{V}_x$, $(-\hat{V}_y)$ and $\hat{V}_z$ are the generators of the SU(1,1) Lie algebra. Equation (A12) can be readily justified by noting that $[\hat{V}_x,\hat{V}_y] = i\hat{V}_z$, which implies the Heisenberg uncertainty relation

$$\langle(\Delta\hat{V}_x)^2\rangle\langle(\Delta\hat{V}_y)^2\rangle \geqslant \tfrac{1}{4}\langle\hat{V}_z\rangle^2.$$

By analogy with the standard quadrature squeezing, sum squeezing occurs when $\min[\langle(\Delta\hat{V}_x)^2\rangle,\langle(\Delta\hat{V}_y)^2\rangle] < \langle\hat{V}_z\rangle/2$, or more generally if Eq. (A12) is satisfied. We note that, in analogy to the principal quadrature squeezing, one can define the principal sum squeezing by minimizing $\langle(\Delta\hat{V}_\phi)^2\rangle$ over ϕ:

$$\min_\phi \langle(\Delta\hat{V}_\phi)^2\rangle < \tfrac{1}{2}\langle\hat{V}_z\rangle. \quad \text{(A13)}$$

Conditions (A12) and (A13) can be easily derived from Criterion 3. In fact, by noting that

$$\langle(\Delta\hat{V}_\phi)^2\rangle = \langle:(\Delta\hat{V}_\phi)^2:\rangle + \tfrac{1}{2}\langle\hat{V}_z\rangle, \quad \text{(A14)}$$

the condition for sum squeezing can equivalently be given by a negative value of the variance $\langle :(\Delta\hat{V}_\phi)^2: \rangle$. On the other hand, by applying Criterion 3 for $\hat{F} = (1, \hat{V}_\phi)$, one obtains

$$d_{\hat{F}}^{(\mathrm{n})} = \begin{vmatrix} 1 & \langle \hat{V}_\phi \rangle \\ \langle \hat{V}_\phi \rangle & \langle :\hat{V}_\phi^2: \rangle \end{vmatrix} = \langle :(\Delta\hat{V}_\phi)^2: \rangle \overset{\mathrm{ncl}}{<} 0, \tag{A15}$$

which is equivalent to Eq. (A12). So it is seen that sum squeezing is a nonclassical effect—in the sense of Criterion 1.

Two-mode sum squeezing can be generalized for any number of modes by defining the following M-mode phase-dependent operator [39]:

$$\hat{\mathcal{V}}_\phi = \frac{1}{2}\left(\mathrm{e}^{-i\phi}\prod_j \hat{a}_j + \mathrm{e}^{i\phi}\prod_j \hat{a}_j^\dagger\right) \tag{A16}$$

satisfying the commutation relation

$$[\hat{\mathcal{V}}_\phi, \hat{\mathcal{V}}_{\phi+\pi/2}] = \frac{i}{2}\hat{C}, \quad \hat{C} = \prod_j (1+\hat{n}_j) - \prod_j \hat{n}_j. \tag{A17}$$

Hereafter $j = 1, \ldots, M$ and we note that $|\langle \hat{C}\rangle| = \langle \hat{C} \rangle$. Thus, multimode sum squeezing along the direction ϕ occurs if

$$\langle (\Delta\hat{\mathcal{V}}_\phi)^2 \rangle < \frac{|\langle \hat{C} \rangle|}{4}. \tag{A18}$$

One can find that

$$\langle (\Delta\hat{\mathcal{V}}_\phi)^2 \rangle = \langle :(\Delta\hat{\mathcal{V}}_\phi)^2: \rangle + \frac{|\langle \hat{C} \rangle|}{4}. \tag{A19}$$

Thus, by applying the nonclassicality Criterion 3 for $\hat{F} = (1, \hat{\mathcal{V}}_\phi)$, we obtain the sum squeezing condition

$$\langle :(\Delta\hat{\mathcal{V}}_\phi)^2: \rangle = d_{\hat{F}}^{(\mathrm{n})} \overset{\mathrm{ncl}}{<} 0, \tag{A20}$$

which is equivalent to condition in Eq. (A18).

4. Difference squeezing

As defined by Hillery [33], a two-mode state exhibits *difference squeezing* in the direction ϕ if

$$\langle (\Delta\hat{W}_\phi)^2 \rangle < \tfrac{1}{2}|\langle \hat{W}_z \rangle|, \tag{A21}$$

where

$$\hat{W}_\phi = \tfrac{1}{2}(\hat{a}_1\hat{a}_2^\dagger e^{i\phi} + \hat{a}_1^\dagger\hat{a}_2 e^{-i\phi}) \tag{A22}$$

and $\hat{W}_z = \frac{1}{2}(\hat{n}_1 - \hat{n}_2)$. The principal difference squeezing can be defined as:

$$\min_\phi \langle (\Delta\hat{W}_\phi)^2 \rangle < \tfrac{1}{2}|\langle \hat{W}_z \rangle|, \tag{A23}$$

in analogy to the principal quadrature squeezing and the principal sum squeezing. Contrary to the $\hat{V}_i$ operators for sum squeezing, operators $\hat{W}_x = \hat{W}(\phi = 0)$, $\hat{W}_y = \hat{W}(\phi = \pi/2)$, and $\hat{W}_z$ are generators of the SU(2) Lie algebra. The uncertainty relation $\langle (\Delta\hat{W}_x)^2\rangle\langle(\Delta\hat{W}_y)^2\rangle \geqslant (1/4)|\langle \hat{W}_z\rangle|^2$ justifies defining difference squeezing by Eq. (A21). One can find that

$$\langle (\Delta\hat{W}_\phi)^2 \rangle = \langle :(\Delta\hat{W}_\phi)^2: \rangle + \tfrac{1}{4}(\langle \hat{n}_1 \rangle + \langle \hat{n}_2 \rangle). \tag{A24}$$

By recalling Criterion 3 for $\hat{F} = (1, \hat{W}_\phi)$, it is seen that

$$d_{\hat{F}}^{(\mathrm{n})} = \langle :(\Delta\hat{W}_\phi)^2: \rangle \overset{\mathrm{ncl}}{<} 0, \tag{A25}$$

in analogy to Eq. (A15). And the condition for sum squeezing, given by Eq. (A21), can be formulated as:

$$d_{\hat{F}}^{(\mathrm{n})} < -\tfrac{1}{2}\min_{i=1,2}\langle \hat{n}_i \rangle. \tag{A26}$$

So states exhibiting difference squeezing are nonclassical. But also states satisfying

$$\tfrac{1}{4}|\langle \hat{n}_1 \rangle - \langle \hat{n}_2 \rangle| \leqslant \langle (\Delta\hat{W}_\phi)^2 \rangle < \tfrac{1}{4}(\langle \hat{n}_1 \rangle + \langle \hat{n}_2 \rangle) \tag{A27}$$

are nonclassical although *not* exhibiting difference squeezing. The first inequality in Eq. (A27) corresponds to condition opposite to squeezing condition given by Eq. (A21).

Criterion 3 can also be applied to the multimode generalization of difference squeezing, which can be defined via the operator [40]:

$$\hat{\mathcal{W}}_\phi = \frac{1}{2}\mathrm{e}^{-i\phi}\prod_{k=1}^{K}\hat{a}_k \prod_{m=K+1}^{M}\hat{a}_m^\dagger + \mathrm{H.c.} \tag{A28}$$

for any $K < M$. For simplicity, hereafter, we skip the limits of multiplication in $\prod_k$ and $\prod_m$. The commutation relation

$$[\hat{\mathcal{W}}_\phi, \hat{\mathcal{W}}_{\phi+\pi/2}] = \frac{i}{2}\hat{C}, \tag{A29}$$

where

$$\hat{C} = \prod_k (1+\hat{n}_k)\prod_m \hat{n}_m - \prod_k \hat{n}_k \prod_m (1+\hat{n}_m), \tag{A30}$$

justifies the choice of the following condition for multimode difference squeezing along the direction ϕ [40]:

$$\langle (\Delta\hat{\mathcal{W}}_\phi)^2 \rangle < \frac{|\langle \hat{C} \rangle|}{4}. \tag{A31}$$

We find that

$$\langle (\Delta\hat{\mathcal{W}}_\phi)^2 \rangle = \langle :(\Delta\hat{\mathcal{W}}_\phi)^2: \rangle + \frac{|\langle \hat{D} \rangle|}{4}, \tag{A32}$$

where

$$\hat{D} = \prod_k (1+\hat{n}_k)\prod_m \hat{n}_m + \prod_k \hat{n}_k \prod_m (1+\hat{n}_m) - 2\prod_{j=1}^{M}\hat{n}_j. \tag{A33}$$

By applying Criterion 3 for $\hat{F} = (1, \hat{\mathcal{W}}_\phi)$, we obtain the following condition for multimode difference squeezing:

$$d_{\hat{F}}^{(\mathrm{n})} = \langle :(\Delta\hat{\mathcal{W}}_\phi)^2: \rangle < \tfrac{1}{4}(|\langle \hat{C} \rangle| - \langle \hat{D} \rangle), \tag{A34}$$

which is equivalent to the original condition, given by Eq. (A31). For states exhibiting difference squeezing, the right-hand side of Eq. (A34) is negative. In fact, if $\langle \hat{C} \rangle > 0$ then

$$\hat{C} - \hat{D} = -2\prod_k \hat{n}_k \left(\prod_m (1+\hat{n}_m) - \prod_m \hat{n}_m\right) < 0, \tag{A35}$$

otherwise

$$\hat{C} - \hat{D} = -2\left(\prod_k (1+\hat{n}_k) - \prod_k \hat{n}_k\right)\prod_m \hat{n}_m < 0. \tag{A36}$$

It is seen that the difference squeezing condition is stronger than the nonclassicality condition $d^{(n)}_{\hat{F}} \overset{\text{ncl}}{<} 0$. This means that states satisfying inequalities

$$\tfrac{1}{4}(|\langle\hat{C}\rangle| - \langle\hat{D}\rangle) \leqslant \langle:(\Delta\hat{\mathcal{W}}_\phi)^2:\rangle < 0 \tag{A37}$$

are nonclassical but *not* exhibiting difference squeezing.

APPENDIX B: UNIFIED DERIVATIONS OF CRITERIA FOR ONE-TIME PHOTON-NUMBER CORRELATIONS

Various criteria for the existence of nonclassical photon-number intermode phenomena in two-mode radiation fields have been proposed (see, e.g., Refs. [3–6,28,31,34]). Here, we give a few examples of such nonclassical phenomena revealed by single-time moments.

1. Sub-Poisson photon-number correlations

The *squeezing* of the sum ($\hat{n}_+ = \hat{n}_1 + \hat{n}_2$) or difference ($\hat{n}_- = \hat{n}_1 - \hat{n}_2$) of photon numbers occurs if

$$\langle:(\Delta\hat{n}_\pm)^2:\rangle < 0, \tag{B1}$$

which can be interpreted as the photon-number sum/difference *sub-Poisson statistics*, respectively [6]. These are nonclassical effects, as can be seen by analyzing the P function:

$$\langle:(\Delta\hat{n}_\pm)^2:\rangle = \int d^2\boldsymbol{\alpha}\; P(\boldsymbol{\alpha},\boldsymbol{\alpha}^*)[(|\alpha_1|^2 \pm |\alpha_2|^2) - \langle\hat{n}_\pm\rangle]^2, \tag{B2}$$

where $\boldsymbol{\alpha} = (\alpha_1,\alpha_2)$. Thus, photon-number squeezing implies the nonpositivity of the P function. The same conclusion can also be drawn by applying Criterion 3 for $\hat{F}_\pm = (1,\hat{n}_\pm)$, which leads to

$$d^{(n)}_{\hat{F}_\pm} = \begin{vmatrix} 1 & \langle\hat{n}_\pm\rangle \\ \langle\hat{n}_\pm\rangle & \langle:\hat{n}^2_\pm:\rangle \end{vmatrix} = \langle:(\Delta\hat{n}_\pm)^2:\rangle \overset{\text{ncl}}{<} 0. \tag{B3}$$

2. Agarwal's nonclassicality criterion

Here, we consider an example of the violation of the CSI for two modes at the same evolution time. Other examples of violations of the CSI for a single mode, but at two different evolution times, are discussed in Appendix C in relation to photon antibunching and hyperbunching.

By considering the violation of the following CSI:

$$\langle:\hat{n}_1^2:\rangle\langle:\hat{n}_2^2:\rangle \overset{\text{cl}}{\geqslant} \langle\hat{n}_1\hat{n}_2\rangle^2, \tag{B4}$$

Agarwal [31] introduced the following nonclassicality parameter:

$$I_{12} = \frac{\sqrt{\langle:\hat{n}_1^2:\rangle\langle:\hat{n}_2^2:\rangle}}{\langle\hat{n}_1\hat{n}_2\rangle} - 1. \tag{B5}$$

Explicitly, the nonclassicality of phenomena described by a negative value of I_{12} is also implied by Criterion 3 for $\hat{F} = (\hat{n}_1,\hat{n}_2)$, which results in

$$d^{(n)}_{\hat{F}} = \begin{vmatrix} \langle:\hat{n}_1^2:\rangle & \langle\hat{n}_1\hat{n}_2\rangle \\ \langle\hat{n}_1\hat{n}_2\rangle & \langle:\hat{n}_2^2:\rangle \end{vmatrix} \overset{\text{ncl}}{<} 0. \tag{B6}$$

3. Lee's nonclassicality criterion

The Muirhead classical inequality [82] is a generalization of the arithmetic-geometric mean inequality. Lee has formulated this inequality as follows [34]

$$D_{12} = \langle:\hat{n}_1^2:\rangle + \langle:\hat{n}_2^2:\rangle - 2\langle\hat{n}_1\hat{n}_2\rangle \overset{\text{cl}}{\geqslant} 0. \tag{B7}$$

The nonclassicality of correlations with a negative value of the parameter D_{12} is readily seen by applying Criterion 3 for $\hat{F} = (\hat{n}_1 - \hat{n}_2) \equiv (\hat{n}_-)$, which yields

$$D_{12} = \langle:\hat{n}_-^2:\rangle \overset{\text{ncl}}{<} 0. \tag{B8}$$

For comparison, let us analyze Criterion 3 for $\hat{F} = (1,\hat{n}_-)$, which leads to

$$d^{(n)}_{\hat{F}} = \langle:\hat{n}_-^2:\rangle - \langle\hat{n}_-\rangle^2 \overset{\text{cl}}{\geqslant} 0. \tag{B9}$$

Clearly

$$D_{12} < 0 \Rightarrow d^{(n)}_{\hat{F}} \overset{\text{ncl}}{<} 0. \tag{B10}$$

Thus, the criterion given by Eq. (B9) detects more nonclassical states than that based on the D_{12} parameter.

Alternatively, a direct application of the relation

$$D_{12} = \int d^2\boldsymbol{\alpha}\; P(\boldsymbol{\alpha},\boldsymbol{\alpha}^*)(|\alpha_1|^2 - |\alpha_2|^2)^2 \overset{\text{ncl}}{<} 0 \tag{B11}$$

also implies the nonpositivity of the P function in some regions of phase space.

APPENDIX C: UNIFIED DERIVATIONS OF CRITERIA FOR TWO-TIME PHOTON-NUMBER CORRELATIONS

Here, we consider the two-time single-mode photon-number nonclassical correlations on examples of photon antibunching and photon hyperbunching.

1. Photon antibunching

The *photon antibunching* [4,5,7,8,43] of a stationary or nonstationary single-mode field can be defined via the two-time second-order intensity correlation function given by

$$\begin{aligned} G^{(2)}(t,t+\tau) &= \langle{}^{\circ}_{\circ}\hat{n}(t)\hat{n}(t+\tau){}^{\circ}_{\circ}\rangle \\ &= \langle\hat{a}^\dagger(t)\hat{a}^\dagger(t+\tau)\hat{a}(t+\tau)\hat{a}(t)\rangle \end{aligned} \tag{C1}$$

or its normalized intensity correlation functions defined as

$$g^{(2)}(t,t+\tau) = \frac{G^{(2)}(t,t+\tau)}{\sqrt{G^{(2)}(t,t)G^{(2)}(t+\tau,t+\tau)}}, \tag{C2}$$

where ${}^{\circ\circ}_{\circ\circ}$ denotes the time order and normal order of field operators. Photon antibunching occurs if $g^{(2)}(t,t)$ is a strict local minimum at $\tau = 0$ for $g^{(2)}(t,t+\tau)$ considered as a function of τ (see, e.g., Refs. [5,37]):

$$g^{(2)}(t,t+\tau) > g^{(2)}(t,t). \tag{C3}$$

Photon bunching occurs if $g^{(2)}(t,t+\tau)$ decreases, while *photon unbunching* appears if $g^{(2)}(t,t+\tau)$ is locally constant.

For *stationary* fields [i.e., those satisfying $G^{(2)}(t,t+\tau) = G^{(2)}(\tau)$ so $g^{(2)}(t,t+\tau) = g^{(2)}(\tau)$], Eq. (C3) reduces to the standard definition of photon antibunching [4,5]:

$$g^{(2)}(\tau) > g^{(2)}(0). \tag{C4}$$

Photon antibunching, defined by Eq. (C3), is a nonclassical effect as it corresponds to the violation of the Cauchy-Schwarz inequality:

$$G^{(2)}(t,t)G^{(2)}(t+\tau,t+\tau) \overset{\text{cl}}{\geqslant} [G^{(2)}(t,t+\tau)]^2. \tag{C5}$$

As shown in Ref. [24], this property follows from Criterion 3 based on the generalized definition of space-time P function, given by (9). In fact, by assuming $\hat{F} = (\hat{n}(t),\hat{n}(t+\tau))$, which leads to

$$\begin{aligned} d_{\hat{F}}^{(\text{n})} &= \begin{vmatrix} \langle {}^{\circ}_{\circ}\hat{n}^2(t){}^{\circ}_{\circ}\rangle & \langle {}^{\circ}_{\circ}\hat{n}(t)\hat{n}(t+\tau){}^{\circ}_{\circ}\rangle \\ \langle {}^{\circ}_{\circ}\hat{n}(t)\hat{n}(t+\tau){}^{\circ}_{\circ}\rangle & \langle {}^{\circ}_{\circ}\hat{n}^2(t+\tau){}^{\circ}_{\circ}\rangle \end{vmatrix} \\ &= \begin{vmatrix} G^{(2)}(t,t) & G^{(2)}(t,t+\tau) \\ G^{(2)}(t,t+\tau) & G^{(2)}(t+\tau,t+\tau) \end{vmatrix} \overset{\text{ncl}}{<} 0. \end{aligned} \tag{C6}$$

2. Photon hyperbunching

Photon hyperbunching [41], also referred to as photon antibunching effect [38], can be defined as:

$$\overline{g}^{(2)}(t,t+\tau) > \overline{g}^{(2)}(t,t), \tag{C7}$$

given in terms of the correlation coefficient [83]

$$\overline{g}^{(2)}(t,t+\tau) = \frac{\overline{G}^{(2)}(t,t+\tau)}{\sqrt{\overline{G}^{(2)}(t,t)\overline{G}^{(2)}(t+\tau,t+\tau)}}, \tag{C8}$$

where the covariance $\overline{G}^{(2)}(t,t+\tau)$ is given by

$$\overline{G}^{(2)}(t,t+\tau) = G^{(2)}(t,t+\tau) - G^{(1)}(t)G^{(1)}(t+\tau), \tag{C9}$$

and $G^{(1)}(t) = \langle\hat{n}(t)\rangle = \langle\hat{a}^\dagger(t)\hat{a}(t)\rangle$ is the light intensity. It is worth noting that, for *stationary* fields, the definitions given by Eqs. (C3) and (C7) are equivalent and equivalent to definitions of photon antibunching based on other normalized correlation functions, e.g.,

$$\tilde{g}^{(2)}(t,t+\tau) = \frac{G^{(2)}(t,t+\tau)}{[G^{(1)}(t)]^2}. \tag{C10}$$

However for *nonstationary* fields, these definitions correspond in general to different photon antibunching effects [37,38,41].

Analogously to Eq. (C3), the photon hyperbunching, defined by Eq. (C7), can occur for nonclassical fields violating the Cauchy-Schwarz inequality:

$$\overline{G}^{(2)}(t,t)\overline{G}^{(2)}(t+\tau,t+\tau) \overset{\text{cl}}{\geqslant} [\overline{G}^{(2)}(t,t+\tau)]^2. \tag{C11}$$

Again, the nonclassicality of this effect can be shown by applying Criterion 3 for the space-time P function, given by (9), assuming $\hat{F} = (\Delta\hat{n}(t),\Delta\hat{n}(t+\tau))$, where $\Delta\hat{n}(t) = \hat{n}(t) - \langle\hat{n}(t)\rangle$. Thus, one obtains

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} \overline{G}^{(2)}(t,t) & \overline{G}^{(2)}(t,t+\tau) \\ \overline{G}^{(2)}(t,t+\tau) & \overline{G}^{(2)}(t+\tau,t+\tau) \end{vmatrix} \overset{\text{ncl}}{<} 0, \tag{C12}$$

which is equivalent to Eq. (C7). Alternatively, by choosing $\hat{F} = (1,\hat{n}(t),\hat{n}(t+\tau))$, one finds

$$d_{\hat{F}}^{(\text{n})} = \begin{vmatrix} 1 & \langle\hat{n}(t)\rangle & \langle\hat{n}(t+\tau)\rangle \\ \langle\hat{n}(t)\rangle & \langle {}^{\circ}_{\circ}\hat{n}^2(t){}^{\circ}_{\circ}\rangle & \langle {}^{\circ}_{\circ}\hat{n}(t)\hat{n}(t+\tau){}^{\circ}_{\circ}\rangle \\ \langle\hat{n}(t+\tau)\rangle & \langle {}^{\circ}_{\circ}\hat{n}(t)\hat{n}(t+\tau){}^{\circ}_{\circ}\rangle & \langle {}^{\circ}_{\circ}\hat{n}^2(t+\tau){}^{\circ}_{\circ}\rangle \end{vmatrix}, \tag{C13}$$

which is equal to the determinant given by Eq. (C12). By comparing Eqs. (C12) and (C13), analogously to Eqs. (A9) and (A10), it is seen the advantage of using polynomial, instead of monomial, functions of moments in $\hat{F}$.

Finally, it is worth noting that the *single-mode sub-Poisson* photon-number statistics, defined by the condition $\langle:(\Delta\hat{n})^2:\rangle < 0$, although also referred to as *photon antibunching*, is an effect different from those defined by Eqs. (C3) and (C7), as shown by examples in Ref. [35].

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