npj | Quantum Information www.nature.com/npjqi

ARTICLE OPEN

Nonclassicality detection from few Fock-state probabilities

Luca Innocenti[1,2,3✉], Lukáš Lachman[1] and Radim Filip[1]

Experimentally certifying the nonclassicality of quantum states in a reliable and efficient way is a challenge that remains both fundamental and daunting. Despite decades of topical research, techniques that can exploit optimally the information available in a given experimental setup are lacking. Here, we introduce a different paradigm to tackle these challenges, that is both directly applicable to experimental realities, and extendible to a wide variety of circumstances. We demonstrate that Klyshko's criteria, which remained a primary approach to tackle nonclassicality for the past 20 years, is a special case of a much more general class of nonclassicality criteria. We provide both analytical results and numerical evidence for the optimality of our approach in several different scenarios of interest for trapped-ion, superconducting circuits, optical and optomechanical experiments with photon-number resolving detectors. This work represents a significant milestone towards a complete characterisation of the nonclassicality detectable from the limited knowledge scenarios faced in experimental implementations.

npj Quantum Information (2022)8:30 ; https://doi.org/10.1038/s41534-022-00538-y

INTRODUCTION

Many fundamental quantum information protocols rely on the nonclassicality of bosonic systems induced by nonlinear phenomena[1,2]. Nonclassical statistics are a crucial resource for quantum sensing[3], as demonstrated by the recent experiments with trapped ions[4,5] and superconducting qubits[6], and more generally for the advancement of quantum information processing[7,8]. Most existing nonclassicality criteria require knowledge of the statistical moments of the boson-number distribution[9–20], which are hard to estimate accurately in experimental platforms such as superconducting-circuits and trapped-ions[21–25], as well as with photon-number-resolving detectors[26–29]. At the same time, not being tailored to the observables that are directly accessible in a given experimental platform, these tests do not make optimal use of the information available. Nonclassicality criteria relying on photon-click statistics[30–35] have similar shortcomings. We here tackle both issues: on the one hand, we devise improved nonclassicality tests that can be directly applied to finite numbers of estimated boson-number probabilities, which is useful to better resolve the different brands of nonclassicality underlying boson-number distributions[24,36–42]. On the other hand, we investigate the ultimate limits of any such test.

While deciding the nonclassicality of an unknown input state is fundamentally impossible with finitely many measurements, we find that, remarkably, in at least some cases of interest it is nonetheless possible to devise criteria that are optimal with respect to a given finite amount of information. Such criteria are very useful in providing definitive answers to precisely which states can be certified as nonclassical in a given experimental scenario. While we focus on the nonclassicality detectable from few Fock-state probabilities, our methodology, based on rather general geometric ideas, can be extended to tackle the nonclassicality of different types of measurements. This approach departs considerably from methods relying on quasiprobability phase-space distributions, which typically rely on complete tomographic information to determine the (non)classicality of a state[43–50]. The two approaches not only provide incomparable results, but also rely on fundamentally different assumptions.

A pioneering step in this direction was taken by D.N. Klyshko[51], who developed nonclassicality criteria—in the form of inequalities for the Fock-state probabilities—satisfied by all classical states. These criteria found numerous applications in both theoretical and experimental contexts[39,52–55]. Similar criteria were also independently formulated in ref. [56]. For many photon and phonon states, Klyshko's inequalities are however still insufficient to detect nonclassicality, and a thorough analysis of their completeness is lacking.

Here, we strengthen Klyshko's methodology, developing criteria to certify nonclassicality from few Fock-state probabilities that are well-suited to experimental implementations. More specifically, we address the open issue of determining, given a vector $\boldsymbol{P} \equiv (P_0, P_1, \ldots, P_n)$ of Fock-state probabilities, whether these probabilities are incompatible with classical states. It is worth stressing that, because knowing the probabilities in a fixed basis is not sufficient to characterize a quantum state, it is possible for a given $\boldsymbol{P}$ to correspond to both classical and non-classical states. Nonetheless, we can assess compatibility with a classical distribution, thus allowing to certify the nonclassicality of a given state. In other words, our approach allows to determine whether nonclassicality is detectable at all from a finite amount of given information. Moreover, we prove that in at least some cases our strengthened criteria are already complete, in the sense that all finite sets of Fock-state probabilities corresponding to nonclassical states are detected as such. A significant advantage of our approach over previous endeavours is our working directly on the Fock-state probabilities, rather than using photon-click statistics or statistical moments. This makes the criteria detector-independent and of broader applicability, in particular in the context of recent optical[57], atomic[58], circuit quantum electrodynamics[59], and optomechanical[60] experiments, and in light of the recent progress in photon-number-resolving detection technology[31,57,61].

While the focus of this paper is on P-nonclassicality[1–3,62], that is, on detecting states whose P function cannot be interpreted as a probability distribution, our approach can be extended to

[1]Department of Optics, Palacký University, 17. Listopadu 12, 771 46 Olomouc, Czech Republic. [2]Centre for Theoretical Atomic, Molecular and Optical Physics, Queen's University Belfast, Belfast BT7 1NN, UK. [3]Dipartimento di Fisica e Chimica - Emilio Segrè, Università degli Studi di Palermo, via Archirafi 36, I-90123 Palermo, Italy.
✉email: luca.innocenti@community.unipa.it

Published in partnership with The University of New South Wales

different notions of nonclassicality, thus paving the way for a similar characterization of non-Gaussianity[35], a crucial resource for quantum computing with bosonic systems.

RESULTS

In this section, we will discuss the extension of Klyshko's criteria[51], and how our geometrical approach that allows to provide definitive answers regarding the question of which nonclassical states can be certified as such in a given scenario where a limited number of observables are accessible.

Klyshko showed that, for all $k \geq 1$, the condition $kP_k^2 > (k+1)P_{k+1}P_{k-1}$ cannot be fulfilled by classical states[51]. Here, we generalize these criteria to make them usable in arbitrary subsets of Fock-state probabilities of the form $\{P_0, P_1, \ldots, P_N\}$. Furthermore, we will show that the nonclassicality criteria involving the unobserved probabilities $\{P_{N+1}, P_{N+2}, \ldots\}$ can be expressed in terms of the observable ones, providing a stronger nonclassicality criterion. We will show that, in the $N = 2$ case, these criteria characterize the set of nonclassical states. This means that our criterion, at least in such special instances, exhausts the amount of information about nonclassicality that can be pried from Fock-state probabilities. Finally, we will showcase applications of our criteria to several classes of experimentally relevant states whose nonclassicality is impervious to alternative methods. The geometrical approach we use to derive our results has several advantages over alternative methods such as those based on the maximization of task-dependent functionals[33]. In the Supplementary Information, we discuss how some of our results could be derived using this approach, which further highlights how the geometric approach might be a preferable venue to tackle the problems studied here. It is worth stressing that our criteria are not directly comparable with standard phase-space-based approaches which rely on the negativity of Wigner or P-function of the full state. Such approaches require full information about the state[43–50], and cannot be directly compared with the results in the partial-knowledge scenario we consider. Same argument applies for momentum-based approaches[9–20]. All these methods share the shortcomings of relying on information about a state that is not easily estimated in many practical scenarios. For the sake of completeness, we present in the Supplementary Information the results obtained using such criteria, in order to highlight the fundamental differences between them.

Boundary of nonclassicality

Let $\mathcal{S} \subset \mathcal{H}$ be the set of quantum states on a single-mode Hilbert space $\mathcal{H}$, and $\mathcal{C}_{\rm coh} \subset \mathcal{S}$ the set of coherent states, that is, of trace-1 projectors of the form $\{|\alpha\rangle\langle\alpha|\}_{\alpha\in\mathbb{C}}$, where $|\alpha\rangle$ denotes a coherent states with average boson number $|\alpha|^2$. Finally, let $\mathcal{C} \subset \mathcal{S}$ denote the set of classical states, that is, the convex hull of $\mathcal{C}_{\rm coh}$. This is the set of states ρ that can be written as $\rho = \int d^2\alpha P(\alpha)|\alpha\rangle\langle\alpha|$ for some probability distribution $P(\alpha)$. Classical states can always be prepared by a classical external driving force on the quantized linear oscillator[63]. As such, their features are explainable via a classical formalism[1].

Given $N \geq 1$, consider the reduced probability space

$$\mathcal{P}_N \equiv \left\{(P_0, P_1, \ldots, P_N) : \sum_{k=0}^{N} P_k \leq 1 \text{ and } P_k \geq 0\right\}. \tag{1}$$

This is the set of vectors which can be part of some larger probability distribution. Denote with $\pi_N : \mathcal{S} \to \mathcal{P}_N$ the natural projection sending each state to its corresponding Fock-state probabilities: $\pi_N(\rho) \equiv (\rho_{kk})_{k=0}^N \in \mathcal{P}_N$. We want to characterize algebraically the projection $\pi_N(\mathcal{C})$ of $\mathcal{C}$ onto the reduced probability space $\mathcal{P}_N$. A crucial observation is that π_N is linear. This implies that convex regions in $\mathcal{S}$ are mapped into convex regions in $\mathcal{P}_N$, and thus in particular $\pi_N(\mathcal{C})$ is convex. Characterizing its boundary $\partial\pi_N(\mathcal{C})$ is therefore sufficient to characterize the whole of $\pi_N(\mathcal{C})$.

Generalizing Klyshko's inequalities

A first investigation of the $N = 2$ case was presented in[33], where nonclassicality criteria using (P_0, P_k) were derived. We summarize and extend these results, discussing the nonclassicality in general spaces of the form (P_n, P_m). We then extend these considerations to bound the possible Fock-state probabilities in arbitrary probability spaces. In particular, we derive criteria in the form of inequalities relating probability tuples $(P_{I_1}, \ldots, P_{I_\ell})$ and $(P_{J_1}, \ldots, P_{J_\ell})$ such that $\sum_i I_i = \sum_i J_i$ and I and J are comparable via majorization. We say that a tuple I is majorized by J, and write $I \preccurlyeq J$, if the sum of the k largest elements of I is smaller than the sum of the k largest elements of J, for all k. We say that I is comparable to J via majorization if either $I \preccurlyeq J$ or $J \preccurlyeq I$. An example of non-comparable tuples is (2, 2, 2, 0) and (3, 1, 1, 1)[64–67]. More precisely, if $I \preccurlyeq J$ and ρ is classical, then the associated probabilities are bound to satisfy

$$\prod_{i=1}^{s} I_i! P_{I_i} \leq \prod_{i=1}^{s} J_i! P_{J_i}, \tag{2}$$

where $s \equiv |I| = |J|$. Each such criterion corresponds to a nonclassicality criterion which can be used when the experimenter is given the corresponding set of Fock-state probabilities.

To prove Eq. (2), we start by defining $Q_k \equiv k!P_k$, so that the statement reads $\prod_{i=1}^s Q_{I_i} \leq \prod_{i=1}^s Q_{J_i}$. Remembering the general identity for products of sums

$$\prod_{i=1}^{n}\sum_{j=1}^{m} a_{ij} = \sum_J \prod_{i=1}^{n} a_{iJ_i}, \tag{3}$$

where the last sum ranges over all multi-indices J of length n, with $J_i \in \{1, \ldots, m\}$ for all $i = 1, \ldots, n$. For any classical state, the probabilities have the form

$$P_k = \sum_\lambda p_\lambda e^{-\lambda}\frac{\lambda^k}{k!}, \tag{4}$$

and thus

$$\prod_i Q_{I_i} = \prod_i \sum_\lambda p_\lambda e^{-\lambda}\lambda^{I_i} = \sum_{\boldsymbol\lambda} p_{\boldsymbol\lambda} e^{|\boldsymbol\lambda|}\boldsymbol\lambda^I, \tag{5}$$

where we used the shorthand notation $p_{\boldsymbol\lambda} \equiv \prod_i p_{\lambda_i}$, $|\boldsymbol\lambda| \equiv \sum_i \lambda_i$, and $\boldsymbol\lambda^I \equiv \prod_i \lambda_i^{I_i}$, and the sum is extended to all possible tuples of values of λ. From the above expression, we see that

$$\prod_i Q_{I_i} - \prod_i Q_{J_i} = \sum_{\boldsymbol\lambda} p_{\boldsymbol\lambda} e^{-|\boldsymbol\lambda|}(\boldsymbol\lambda^I - \boldsymbol\lambda^J). \tag{6}$$

The conclusion then follows from *Miurhead's inequalities*[64,68]. More details are provided in the Supplementary Information.

In particular, when I, J have length 3, we get inequalities involving triples of probabilities: for all $0 \leq n \leq m \leq k$, classical states are bound to satisfy:

$$(m!P_m)^{k-n} \leq (n!P_n)^{k-m}(k!P_k)^{m-n}. \tag{7}$$

Violation of Eq. (7) thus certifies nonclassicality. For $n = N-1, m = N$ and $k \geq N$, defining $Q_k \equiv k!P_k$, we have $Q_N^{k-N+1} \leq Q_{N-1}^{k-N}Q_k$, and thus

$$k!P_k \geq \frac{Q_{N-1}^N}{Q_N^{N-1}}\left(\frac{Q_N}{Q_{N-1}}\right)^k, \ \forall k \geq N-1. \tag{8}$$

Using this in conjunction with the normalization condition $\sum_k P_k = 1$ we get

$$\sum_{k=0}^{N-2} P_k + \frac{Q_{N-1}^N}{Q_N^{N-1}}\sum_{k=N-1}^{\infty}\frac{1}{k!}\left(\frac{Q_N}{Q_{N-1}}\right)^k \leq 1. \tag{9}$$

Using the Taylor expansion of the exponential function to write $\sum_{k=N-1}^{\infty}\frac{x^k}{k!}=e^x-\sum_{k=0}^{N-2}\frac{x^k}{k!}$, we then conclude that all classical states must satisfy the inequality

$$\sum_{k=0}^{N-2}P_k+\frac{Q_{N-1}^N}{Q_N^{N-1}}\left[e^{\frac{Q_N}{Q_{N-1}}}-\sum_{k=0}^{N-2}\frac{(Q_N/Q_{N-1})^k}{k!}\right]\le 1. \tag{10}$$

Together with the standard Klyshko conditions in the form $Q_k^2\le Q_{k-1}Q_{k+1}$, Eq. (10), defines a closed region $\mathcal{D}_N\subset\mathcal{P}_N$ containing $\pi_N(\mathcal{C})$. Any probability vector $\boldsymbol{P}\notin\mathcal{D}_N$ is certifiably nonclassical. In the rest of the paper, we will refer to condition (10) as $\mathcal{K}_{\infty,N}$, and to the Klyshko condition $Q_k^2\le Q_{k-1}Q_{k+1}$ as $\mathcal{K}_k$. We will also use $\mathcal{K}_\infty$ to refer more generally to criteria of the type $\mathcal{K}_{\infty,N}$ for some N.

Let us remark two additional facts:

1. Having access to a finite set of probabilities $(P_0,\dots,P_{N-1})$, there are always nonclassical states that are not detectable by any criterion. For example, consider $\rho^{(N)}\equiv p\rho_{\rm cl}+(1-p)|N\rangle\langle N|$ for some classical state $\rho_{\rm cl}$. Having only access to the first N probabilities amounts to working with the reduced distribution $\pi_{N-1}(\rho^{(N)})=p\pi_{N-1}(\rho_{\rm cl})$. Being $\rho_{\rm cl}$ classical, as we discussed previously, $\pi_{N-1}(\rho_{\rm cl})$ satisfies all the inequalities $\mathcal{K}_\infty$ and $\mathcal{K}_k$. The scaled probability vector $p\pi_{N-1}(\rho_{\rm cl})$ is then also bound to satisfy the Klyshko-like inequalities $\mathcal{K}_k$, as these are scale-invariant. It is then also easy to verify that if $\mathcal{K}_{\infty,N-1}$ is satisfied for $\pi_{N-1}(\rho_{\rm cl})$, then it must also be satisfies for $p\pi_{N-1}(\rho_{\rm cl})$ for any $0<p<1$.
2. The $\mathcal{K}_\infty$ inequalities are a necessary addition to fully expoit the knowledge encoded in the Fock-number probability distributions. Indeed, there are always nonclassical states undetected by the Klyshko-like inequalities. For example, knowing any set $(P_0,\dots,P_N)$, the Fock state $|N\rangle$ can be seen to satisfy all inequalities of type $\mathcal{K}_k$, but it violates $\mathcal{K}_{\infty,N}$.

The fundamental question that remains to be addressed is whether the inequalities of the form $\mathcal{K}_k$ and $\mathcal{K}_\infty$ exhaust the information about nonclassicality encoded in Fock-state probabilities. This amounts to asking whether a probability vector sastisfying all the inequalities (those usable given a finite set of probabilities), implies the existence of a classical state resulting in said probabilities. In other words, we want to know whether satisfying all relevant inequalities certifies compatibility with some classical state, which is the most one can ask for in this scenario.

Nonclassicality in (P_0,P_1,P_2)

To analyse the applicability of the conditions $\mathcal{K}_{\infty,N}$ beyond[51], we study what nonclassical states can be detected when only the first three Fock-state probabilities are known. We will find that, remarkably, the nonclassicality of a state is completely captured by only two algebraic inequalities.

Let us denote with $\mathcal{K}_1$ the region:

$$\mathcal{K}_1\equiv\{(P_0,P_1,P_2)\in\mathcal{P}_2:P_1^2=2P_0P_2\}, \tag{11}$$

and with $\mathcal{K}_{\infty,2}$ the set of points satisfying Eq. (10) with $N=2$, that is, the probability vectors $(P_0,P_1,P_2)\in\mathcal{P}_2$ such that $P_0+\frac{P_1^2}{2P_2}\left[\exp(\frac{2P_2}{P_1})-1\right]=1$. The associated nonclassicality criteria are then

$$P_1^2>2P_0P_2, \tag{12}$$

$$P_0+\frac{P_1^2}{2P_2}\left[\exp\left(\frac{2P_2}{P_1}\right)-1\right]>1. \tag{13}$$

The notation $\mathcal{K}_1^{\gtrless}$ and $\mathcal{K}_{\infty,2}^{\gtrless}$ will be used to denote the sets obtained by replacing the equality in these definitions with the corresponding inequality sign (e.g. $\mathcal{K}_1^{\ge}$ is the set of points such that $P_1^2\ge 2P_0P_2$, while $\mathcal{K}_1^{<}$ is the set of points such that $P_1^2<2P_0P_2$). We will prove in this section that $\pi_2(\mathcal{C})=\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}^{\le}$, that is, that Eqs. (12) and (13) are necessary and sufficient conditions for a state being detectable as nonclassical when only knowledge of the first three Fock-state probabilities is given. It is worth stressing that these nonclassicality criteria are strictly stronger than previously reported criteria using pairs of Fock-state probabilities[33].

We already showed that all classical states are contained in $\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}^{\le}$. To prove that the inequalities provide a necessary and sufficient condition for compatibility with classical states, we need to show that any probability vector in $\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}^{\le}$ is compatible with a classical state. For the purpose, we will show that any $\boldsymbol{P}$ inside this region can be written as convex combination of probability vectors compatible with classical states. In other words, we want to find the mixture of coherent states corresponding to a given triple of probabilities $\boldsymbol{P}\equiv(P_0,P_1,P_2)\in\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}^{\le}$. To achieve this, we will (1) show that $\boldsymbol{P}$ is a convex mixture of the origin and a point $\boldsymbol{P}_\infty\in\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}$; (2) show that $\boldsymbol{P}_\infty$ can be written as convex combination of (1, 0, 0) (the point corresponding to the vacuum state) and an element of $\mathcal{K}_1\cap\mathcal{K}_{\infty,2}$; (3) show that all vectors in $\mathcal{K}_1\cap\mathcal{K}_{\infty,2}$ are compatible with coherent states. This will allow us to conclude that $\boldsymbol{P}$ is compatible with a convex combination of coherent states.

Let $\boldsymbol{P}\in\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}^{\le}$ be an arbitrary point not satisfying the nonclassicality criteria (Eqs. (12) and (13)). Define the quantities $K_1(\boldsymbol{P})\equiv P_1^2-2P_0P_2$ and $K_{\infty,2}(\boldsymbol{P})\equiv P_0+\frac{P_1^2}{2P_2}[\exp(2P_2/P_1)-1]$. Note that, upon rescaling $\boldsymbol{P}\to\epsilon\boldsymbol{P}$, the sign of K_1 is invariant, and $K_{\infty,2}\to\epsilon K_{\infty,2}$. We can therefore always find $\epsilon\ge 1$ such that $\boldsymbol{P}'\equiv\epsilon\boldsymbol{P}\in\mathcal{K}_1^{\le}\cap\mathcal{K}_{\infty,2}$. We can thus write $\boldsymbol{P}$ as a convex combination of $\boldsymbol{P}'$ and the origin in probability space, $\boldsymbol{0}\equiv(0,0,0)$, as $\boldsymbol{P}=1/\epsilon\boldsymbol{P}'+(1-1/\epsilon)\boldsymbol{0}$. Note that $\boldsymbol{0}$ is the probability vector generated by a coherent state in the limit of infinite average boson number, and is thus classical. It now remains to prove that $\boldsymbol{P}'$ is also classical to conclude that $\boldsymbol{P}$ is. For the purpose, consider convex combinations of $\boldsymbol{P}'$ and $\boldsymbol{e}_0\equiv(1,0,0)\equiv\pi_2(|0\rangle\langle 0|)$, and notice that

$$\begin{aligned}K_{\infty,2}(p\boldsymbol{P}'+(1-p)\boldsymbol{e}_0)&=pK_{\infty,2}(\boldsymbol{P}')+(1-p)=1,\\ K_1(p\boldsymbol{P}'+(1-p)\boldsymbol{e}_0)&=p^2K_1(\boldsymbol{P}')-2p(1-p)P_2'.\end{aligned} \tag{14}$$

Solving for the $p\neq 0$ such that $K_1=0$, we find

$$p=\frac{2P_2'}{(P_1')^2}e^{-2P_2'/P_1'}\ge 1, \tag{15}$$

where we used $K_{\infty,2}(\boldsymbol{P}')=1$ and $K_1(\boldsymbol{P}')\le 0$. This means that there is some $\boldsymbol{P}''\in\mathcal{K}_1\cap\mathcal{K}_{\infty,2}$ such that $\boldsymbol{P}'=\frac{p-1}{p}\boldsymbol{e}_0+\frac{1}{p}\boldsymbol{P}''$. To conclude, we now only need to show that $\boldsymbol{P}''$ is compatible with a coherent state. By definition of $\mathcal{K}_1\cap\mathcal{K}_{\infty,2}$, the elements of $\boldsymbol{P}''$ satisfy

$$\frac{P_1''^2}{2P_2''}e^{2P_2''/P_1''}=P_0''e^{P_1''/P_0''}=1. \tag{16}$$

We finally note that, for any value of P_0'', these conditions uniquely determine P_1'' and P_2'', and that a coherent state with average boson number $\mu=-\log P_0$ produces these probabilities.

It is worth remarking that the above reasoning not only proves that the given inequalities characterize the boundary of classical states in $\mathcal{P}_2$, but also provides a constructive method to find classical states compatible with an observed (not nonclassical) probability distribution.

Applications: Fock and squeezed states

A class of states benefiting from the $\mathcal{K}_\infty$ criteria are Fock states. The Fock state $|1\rangle\equiv a^\dagger|0\rangle$ clearly satisfies $P_1^2>2P_0P_2$, and is therefore detected as nonclassical by $\mathcal{K}_1$. More generally, convex mixtures of $|0\rangle$ and $|1\rangle$ are all detected as nonclassical by $\mathcal{K}_1$ but not by $\mathcal{K}_{\infty,2}$, as also seen in Fig. 1. On the other hand, $|2\rangle=\frac{1}{\sqrt{2}}a^{\dagger 2}|0\rangle$ is detected as nonclassical by $\mathcal{K}_{\infty,2}$ but not by $\mathcal{K}_1$.

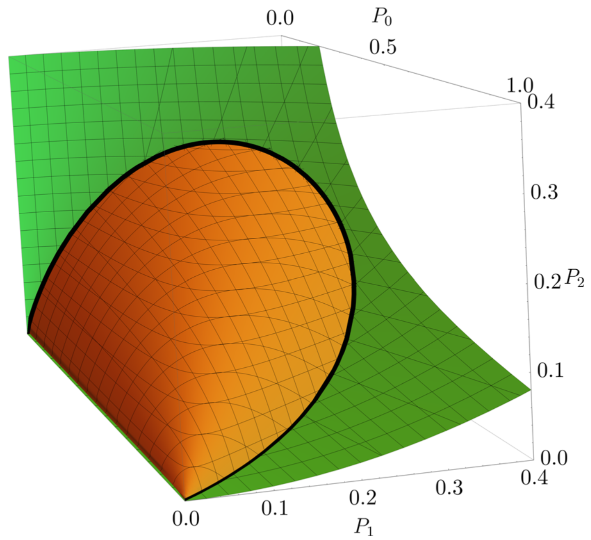

Fig. 1 Classical set in the (P_0, P_1, P_2) space. The orange (upper) surface is the set of points on $\mathcal{K}_\infty$, while the green (lower) surface the set of points on $\mathcal{K}_1$. The black line is the set of coherent states. We notice that the upper surface can be generated as the set of lines going from $(1, 0, 0)$ to the coherent states, while the lower surface as the set of lines going from $(0, 0, 0)$ to the coherent states. All the states with probabilities lying above the upper surface will satisfy Klyshko's inequalities, and can therefore be detected as nonclassical only using Eq. (13).

Consider now attenuated Fock states, that is, states of the form $\mathcal{E}_T(|k\rangle\langle k|)$ with $|k\rangle$ Fock states and $\mathcal{E}_T$ the channel corresponding to attenuation through a beamsplitter with transmittivity $T\in[0,1]$ (thus, in particular, $\mathcal{E}_1(\rho)=\rho$ and $\mathcal{E}_0(\rho)=\mathrm{Tr}(\rho)|0\rangle\langle 0|$ for all ρ). In these cases, we find that $\mathcal{E}_T(|k\rangle\langle k|)$ is, in principle, detected as nonclassical by both $\mathcal{K}_1$ and $\mathcal{K}_{\infty,2}$, for all $T\in[0,1]$ and $k\in\mathbb{N}$. However, the criteria detect this nonclassicality very differently: it is harder to detect the nonclassicality with $\mathcal{K}_1$ for T closer to 1, while $\mathcal{K}_{\infty,2}$ makes it easier in this regime, and viceversa for smaller values of T. A Fock state such as $|2\rangle$ sits on the boundary of the classical region, and is therefore undetectable as nonclassical with finite statistics with $\mathcal{K}_1$, which is why these results are consistent with our previous statement that $|2\rangle$ can only be detected as nonclassical with $\mathcal{K}_{\infty,2}$. This hardness for T approaching unity increases for higher Fock states $|k\rangle$, as shown in the Supplementary Information. More generally, $\mathcal{K}_1$ cannot certify the nonclassicality of convex mixtures of $|2\rangle$ and $|0\rangle$, which is however revealed by $\mathcal{K}_{\infty,2}$. This suggests squeezed states as another class benefiting from our extended criteria.

In the Supplementary Information we show that many squeezed thermal states[69] also require $\mathcal{K}_{\infty,2}$ to be detected as nonclassical.

Applications: Boson-added noisy states

Photon- and phonon-added coherent states[70–72] are defined as $|\alpha,\ell\rangle\equiv C_{\alpha,\ell}a^{\dagger\ell}|\alpha\rangle$ with $C_{\alpha,\ell}$ normalization constants. The associated Fock-state distribution equals that obtained adding single bosons to Poissonian noise with average number $\mu=|\alpha|^2$, here denoted ρ_μ. The introduced criteria provide enhanced predictive power also for these highly noisy states ρ_μ. For example, for $\ell=2$, $\mathcal{K}_1$ does not predict nonclassicality with P_0, P_1, P_2, but $\mathcal{K}_{\infty,2}$ does. The same holds for probabilistic boson addition. Consider, e.g. states of the form $p\,a^\dagger\rho_\mu a+(1-p)\rho_\mu$. We find that using $\mathcal{K}_\infty$ criteria allows to detect nonclassicality more efficiently, as shown in Fig. 2. More details are found in the Supplementary Information.

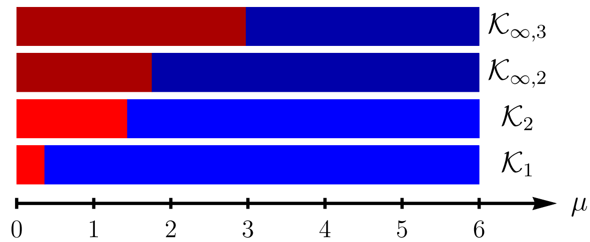

Fig. 2 Nonclassicality of boson-added states $p\,a^\dagger\rho_\mu a+(1-p)\rho_\mu$ with $p=0.5$. For each μ, we highlight whether the different criteria detect the corresponding probability distribution as nonclassical (red) or not (blue). Note how restricting to the first three Fock-state probabilities, when only $\mathcal{K}_1$ and $\mathcal{K}_{\infty,2}$ are accessible, nonclassicality is certified only up to $\mu\sim 1.7$. On the other hand, knowing P_3, nonclassicality is certifiable up to $\mu\sim 3$, thanks to $\mathcal{K}_{\infty,3}$.

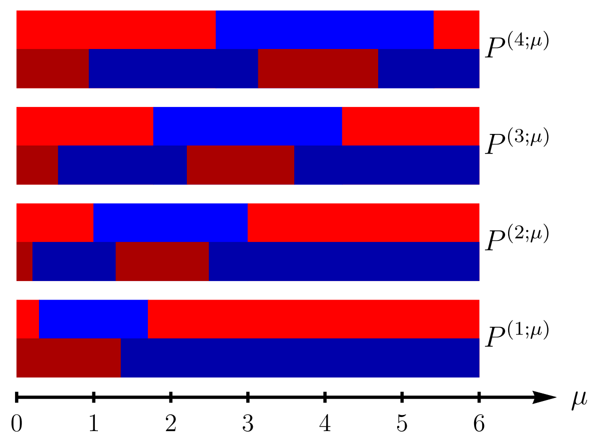

Fig. 3 Nonclassicality of noisy Fock states. Notation is as in Fig. 2. We highlight the regions of nonclassicality detected by $\mathcal{K}_1$ (upper light red) and $\mathcal{K}_{\infty,2}$ (lower dark red) for noisy Fock states statistics $P^{(k;\mu)}$, for $k=1,2,3,4$. For example, we see that $\mu=1.5$ corresponds to a classical statistics for $P^{(1;\mu)}$, a nonclassical one detected by $\mathcal{K}_{\infty,2}$ for $P^{(2;\mu)}$, and a nonclassical one also for $P^{(3;\mu)}$ and $P^{(4;\mu)}$, now detected by $\mathcal{K}_1$. Further details can be found in Supplementary Figs. 6 and 7.

Applications: Noisy Fock states

Consider now displaced Fock states, $|\alpha;k\rangle=D(\alpha)|k\rangle$, obtained applying the displacement operator $D(\alpha)$ to a Fock state $|k\rangle$[73]. Averaging over the phases of α, these produce the same Fock-state probabilities $(P_j^{(k;\mu)})_j$ as Fock states with added Poissonian noise, and model states produced in realistic experimental conditions, where the displacement operator causes Fock states higher than $|k\rangle$ to contribute. As shown in Fig. 3, $\mathcal{K}_\infty$ criteria increase the predictive power when few Fock-state probabilities are known. For example, for $k=1$, when P_0, P_1, P_2 are known, $\mathcal{K}_1$ does not certify nonclassicality for $0.29\lesssim\mu\lesssim 1.71$, but $\mathcal{K}_{\infty,2}$ does for $\mu\lesssim 1.35$. This means that $\mathcal{K}_{\infty,2}$ allows to detect nonclassical states in regimes in which $\mathcal{K}_1$ is not sufficient. Another striking feature emerging from Fig. 3 is that adding more noise can make it easier to detect nonclassicality, as highlighted by the presence of bright red regions for large values of μ. This remains the case even if, instead of simply increasing the average boson number of the added Poissonian noise, we add incoherent noise to the state. We find that this can also make the nonclassicality of a distribution easier to detect. More details are found in the Supplementary Information.

DISCUSSION

We showed that Klyshko's criteria are a special case of a broader class of nonclassicality criteria. Leveraging this result we found that, when only few Fock-state probabilities are known, these enhanced criteria grant additional insight into nonclassical properties of boson statistics, even in realistic experimental conditions. Such criteria are pivotal to deepen our understanding of nonclassical phenomena and uncover additional resources for quantum technologies. Our method is directly applicable to trapped-ion[4,5], superconducting-circuit[6], and optical experiments with photon-number resolving detectors[31,57,61]. More specifically, the application of the proposed methodology to a given experimental scenario is completely straightforward, only requiring to verify whether the measured quantities satisfy a finite set of algebraic inequalities.

We proved the optimality of our improved criteria with respect to the first three Fock-state probabilities, and discussed a number of example applications of the criteria for several classes of states of interest, including boson-added thermal states, noisy Fock states, and thermal and Fock states. Others, such as thermal states, are also considered and discussed in the Supplementary Information. We remark that even in cases in which the $\mathcal{K}_\infty$ criteria do not provide additional predictive capabilities, as is the case for example for some classes of noisy Fock states, discussed in the section "Applications: Noisy Fock states" and in the Supplementary Information, and boson-added thermal states, discussed in the Supplementary Information. We stress that, even in these cases, our analysis is useful allowing to conclude that the nonclassicality certification problem, in some circumstances, is simply unsolvable with the information given.

The optimality of the proposed criteria in higher-dimensional slices of probability space remains a stimulating open question, which if solved would provide further insight into the nonclassicality of boson statistics. Another interesting aspect emerging from a combination of this approach with the methodology of Filip and Lachman[33], is how adding noise to a state, which is generally an easy operation, can make it easier to detect the nonclassicality of the state from its Fock-state distribution. Our results, paired with modern optimization techniques, pave the way to a complete characterization of the nonclassicality accessible from finite sets of measurable quantities.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: 3 May 2021; Accepted: 26 January 2022;
Published online: 18 March 2022

REFERENCES

1. Glauber, R. J. Coherent and incoherent states of the radiation field. *Phys. Rev.* **131**, 2766–2788 (1963).
2. Sudarshan, E. C. G. Equivalence of semiclassical and quantum mechanical descriptions of statistical light beams. *Phys. Rev. Lett.* **10**, 277–279 (1963).
3. Tan, K. C. & Jeong, H. Nonclassical light and metrological power: an introductory review. *AVS Quantum Sci.* **1**, 014701 (2019).
4. Zhang, J. et al. Noon states of nine quantized vibrations in two radial modes of a trapped ion. *Phys. Rev. Lett.* **121**, 160502 (2018).
5. Wolf, F. et al. Motional fock states for quantum-enhanced amplitude and phase measurements with trapped ions. *Nat. Commun.* **10**, 1–8 (2019).
6. Wang, W. et al. Heisenberg-limited single-mode quantum metrology in a superconducting circuit. *Nat. Commun.* **10**, 1–6 (2019).
7. Gao, Y. Y. et al. Entanglement of Bosonic modes through an engineered exchange interaction. *Nature* **566**, 509–512 (2019).
8. Gan, H. C. J., Maslennikov, G., Tseng, K.-W., Nguyen, C. & Matsukevich, D. Hybrid quantum computing with conditional beam splitter gate in trapped ion system. *Phys. Rev. Lett.* **124**, 170502 (2020).
9. Agarwal, G. S. & Tara, K. Nonclassical character of states exhibiting no squeezing or sub-Poissonian statistics. *Phys. Rev. A* **46**, 485–488 (1992).
10. Richter, T. & Vogel, W. Nonclassicality of quantum states: a hierarchy of observable conditions. *Phys. Rev. Lett.* **89**, 283601 (2002).
11. Miranowicz, A., Bartkowiak, M., Wang, X., Liu, Y.-x & Nori, F. Testing nonclassicality in multimode fields: a unified derivation of classical inequalities. *Phys. Rev. A* **82**, 013824 (2010).
12. Sperling, J., Vogel, W. & Agarwal, G. S. Sub-binomial light. *Phys. Rev. Lett.* **109**, 093601 (2012).
13. Peřina, J., Arkhipov, I. I., Michálek, V. & Haderka, O. Nonclassicality and entanglement criteria for bipartite optical fields characterized by quadratic detectors. *Phys. Rev. A* **96**, 043845 (2017).
14. Peřina, J., Michálek, V. & Haderka, O. Higher-order sub-Poissonian-like nonclassical fields: Theoretical and experimental comparison. *Phys. Rev. A* **96**, 033852 (2017).
15. Peřina, J., Haderka, V. & Michálek, O. Simultaneous observation of higher-order non-classicalities based on experimental photocount moments and probabilities. *Sci. Rep.* **9**, 1–8 (2019).
16. Kimble, H. J., Dagenais, M. & Mandel, L. Photon antibunching in resonance fluorescence. *Phys. Rev. Lett.* **39**, 691–695 (1977).
17. Grangier, P., Roger, G. & Aspect, A. Experimental evidence for a photon anticorrelation effect on a beam splitter: a new light on single-photon interferences. *Europhys. Lett.* **1**, 173–179 (1986).
18. Grünwald, P. Effective second-order correlation function and single-photon detection. *N. J. Phys.* **21**, 093003 (2019).
19. Grünwald, P. Nonquantum information gain from higher-order correlation functions. *Phys. Rev. Res.* **2**, 023147 (2020).
20. Chavez-Mackay, J. R., Grünwald, P. & Rodríguez-Lara, B. M. Estimating the single-photon projection of low-intensity light sources. *Phys. Rev. A* **101**, 053815 (2020).
21. Um, M. et al. Phonon arithmetic in a trapped ion system. *Nat. Commun.* **7**, 1–7 (2016).
22. Wang, W. et al. Converting quasiclassical states into arbitrary fock state superpositions in a superconducting circuit. *Phys. Rev. Lett.* **118**, 223604 (2017).
23. Kienzler, D. et al. Quantum harmonic oscillator state control in a squeezed fock basis. *Phys. Rev. Lett.* **119**, 033602 (2017).
24. Chu, Y. et al. Creation and control of multi-phonon fock states in a bulk acoustic-wave resonator. *Nature* **563**, 666–670 (2018).
25. Gely, M. F. et al. Observation and stabilization of photonic fock states in a hot radio-frequency resonator. *Science* **363**, 1072–1075 (2019).
26. Kardynał, B. E., Yuan, Z. L. & Shields, A. J. An avalanche photodiode-based photon-number-resolving detector. *Nat. Photonics* **2**, 425–428 (2008).
27. Divochiy, A. et al. Superconducting nanowire photon-number-resolving detector at telecommunication wavelengths. *Nat. Photonics* **2**, 302–306 (2008).
28. Namekata, N. et al. Non-gaussian operation based on photon subtraction using a photon-number-resolving detector at a telecommunications wavelength. *Nat. Photonics* **4**, 655–660 (2010).
29. Endo, M. et al. Quantum detector tomography of a superconducting nanostrip photon-number-resolving detector. *Opt. Express* **29**, 11728 (2021).
30. Rigovacca, L., Di Franco, C., Metcalf, B. J., Walmsley, I. A. & Kim, M. S. Nonclassicality criteria in multiport interferometry. *Phys. Rev. Lett.* **117**, 213602 (2016).
31. Sperling, J. et al. Detector-independent verification of quantum light. *Phys. Rev. Lett.* **118**, 163602 (2017).
32. Sperling, J. et al. Identification of nonclassical properties of light with multiplexing layouts. *Phys. Rev. A* **96**, 013804 (2017).
33. Filip, R. & Lachman, L. Hierarchy of feasible nonclassicality criteria for sources of photons. *Phys. Rev. A* **88**, 043827 (2013).
34. Lachman, L. & Filip, R. Criteria for single photon sources with variable nonclassicality threshold. *N. J. Phys.* **21**, 083012 (2019).
35. Lachman, L., Straka, I., Hloušek, J., Ježek, M. & Filip, R. Faithful hierarchy of genuine n-photon quantum non-gaussian light. *Phys. Rev. Lett.* **123**, 043601 (2019).
36. Zavatta, A., Viciani, S. & Bellini, M. Quantum-to-classical transition with single-photon-added coherent states of light. *Science* **306**, 660–662 (2004).
37. Zavatta, A., Parigi, V. & Bellini, M. Experimental nonclassicality of single-photon-added thermal light states. *Phys. Rev. A* **75**, 052106 (2007).
38. Slodička, L., Marek, P. & Filip, R. Deterministic nonclassicality from thermal states. *Opt. Express* **24**, 7858 (2016).
39. Marek, P., Lachman, L., Slodička, L. & Filip, R. Deterministic nonclassicality for quantum-mechanical oscillators in thermal states. *Phys. Rev. A* **94**, 013850 (2016).
40. Li, J., Gröblacher, S., Zhu, S.-Y. & Agarwal, G. S. Generation and detection of non-gaussian phonon-added coherent states in optomechanical systems. *Phys. Rev. A* **98**, 011801 (2018).
41. Ding, S., Maslennikov, G., Hablützel, R. & Matsukevich, D. Quantum simulation with a trilinear hamiltonian. *Phys. Rev. Lett.* **121**, 130502 (2018).
42. Podhora, L. et al. Unconditional accumulation of nonclassicality in a single-atom mechanical oscillator. Preprint at https://arxiv.org/abs/2004.12863 (2020).

43. Kiesel, T., Vogel, W., Parigi, V., Zavatta, A. & Bellini, M. Experimental determination of a nonclassical Glauber–Sudarshan *P* function. *Phys. Rev. A* **78**, 021804 (2008).
44. Lvovsky, A. I. & Raymer, M. G. Continuous-variable optical quantum-state tomography. *Rev. Mod. Phys.* **81**, 299–332 (2009).
45. Kiesel, T., Vogel, W., Bellini, M. & Zavatta, A. Nonclassicality quasiprobability of single-photon-added thermal states. *Phys. Rev. A* **83**, 032116 (2011).
46. Kühn, B. & Vogel, W. Quantum non-gaussianity and quantification of nonclassicality. *Phys. Rev. A* **97**, 053823 (2018).
47. Sperling, J. & Walmsley, I. A. Quasiprobability representation of quantum coherence. *Phys. Rev. A* **97**, 062327 (2018).
48. Tan, K. C., Choi, S. & Jeong, H. Negativity of quasiprobability distributions as a measure of nonclassicality. *Phys. Rev. Lett.* **124**, 110404 (2020).
49. Bohmann, M. & Agudelo, E. Phase-space inequalities beyond negativities. *Phys. Rev. Lett.* **124**, 133601 (2020).
50. Sperling, J. et al. Detector-agnostic phase-space distributions. *Phys. Rev. Lett.* **124**, 013605 (2020).
51. Klyshko, D. N. Observable signs of nonclassical light. *Phys. Lett. A* **213**, 7–15 (1996).
52. Lee, C. T. Application of Klyshko's criterion for nonclassical states to the micromaser pumped by ultracold atoms. *Phys. Rev. A* **55**, 4449–4453 (1997).
53. Waks, E., Sanders, B. C., Diamanti, E. & Yamamoto, Y. Highly nonclassical photon statistics in parametric down-conversion. *Phys. Rev. A* **73**, 033814 (2006).
54. Wakui, K. et al. Ultrabroadband direct detection of nonclassical photon statistics at telecom wavelength. *Sci. Rep.* **4**, 1–8 (2014).
55. Kono, S. et al. Nonclassical photon number distribution in a superconducting cavity under a squeezed drive. *Phys. Rev. Lett.* **119**, 023602 (2017).
56. Simon, R., Selvadoray, M., Arvind & Mukunda, N. Nonclassicality and the concept of local constraints on the photon number distribution. Preprint at https://arxiv.org/abs/quant-ph/9708038 (1997).
57. Harder, G. et al. Single-mode parametric-down-conversion states with 50 photons as a source for mesoscopic quantum optics. *Phys. Rev. Lett.* **116**, 143601 (2016).
58. Hacker, B. et al. Deterministic creation of entangled atom-light Schrödinger-cat states. *Nat. Photonics* **13**, 110–115 (2019).
59. Sokolov, A. M. & Wilhelm, F. K. Superconducting detector that counts microwave photons up to two. *Phys. Rev. Appl.* **14**, 064063 (2020).
60. Hong, S. et al. Hanbury brown and twiss interferometry of single phonons from an optomechanical resonator. *Science* **358**, 203–206 (2017).
61. Tiedau, J. et al. Scalability of parametric down-conversion for generating higher-order fock states. *Phys. Rev. A* **100**, 041802 (2019).
62. Albarelli, F., Ferraro, A., Paternostro, M. & Paris, M. G. A. Nonlinearity as a resource for nonclassicality in anharmonic systems. *Phys. Rev. A* **93**, 032112 (2016).
63. Scully, M. O. & Zubairy, M. S. *Quantum Optics* (Cambridge University Press, 1997).
64. Hardy, G. H., Littlewood, J. E. & Pólya, G. In *Inequalities* (eds Hardy, G. H., Littlewood, J. E. & Pólya, G.) (University Press, 1952).
65. Muirhead, R. F. Some methods applicable to identities and inequalities of symmetric algebraic functions of n letters. *Proc. Edinb. Math. Soc.* **21**, 144–162 (1902).
66. Marshall, A. W., Olkin, I. & Arnold, B. C. *Inequalities: Theory of Majorization and its Applications*, Vol. 143 (Springer, 1979).
67. Bhatia, R. *Matrix Analysis*, Vol. 169 (Springer Science & Business Media, 2013).
68. Cvetkovski, Z. Schur's inequality, Muirhead's inequality and Karamata's inequality. *Inequalities* 121–132. https://link.springer.com/chapter/10.1007/978-3-642-23792-8_12 (2012).
69. Kim, M. S., de Oliveira, F. A. M. & Knight, P. L. Properties of squeezed number states and squeezed thermal states. *Phys. Rev. A* **40**, 2494–2503 (1989).
70. Agarwal, G. S. & Tara, K. Nonclassical properties of states generated by the excitations on a coherent state. *Phys. Rev. A* **43**, 492–497 (1991).
71. Domínguez-Serna, F. A., Mendieta-Jimenez, F. J. & Rojas, F. Entangled photon-added coherent states. *Quantum Inf. Process.* **15**, 3121–3136 (2016).
72. Gard, B. T. et al. Photon added coherent states: nondeterministic, noiseless amplification in quantum metrology. Preprint at https://arxiv.org/abs/1606.09598 (2016).
73. Bužek, V., Lai, W. & Knight, P. Displaced number states. In *Quantum Aspects of Optical Communications*, 295–304 (Springer, 1991).

ACKNOWLEDGEMENTS

The authors acknowledge helpful discussions with Mauro Paternostro and Alessandro Ferraro. L.L. and R.F. acknowledge project 21-13265X of the Czech Science Foundation. L.I. acknowledges support from *Fondazione Angelo della Riccia* and support from MEYS CR and European Union's Horizon 2020 (2014–2020) research and innovation framework programme under grant agreement No. 731473 (project 8C20002 ShoQC). Project ShoQC has received funding from the QuantERA ERA-NET Cofund in Quantum Technologies implemented within the European Union's Horizon 2020 Programme.

AUTHOR CONTRIBUTIONS

The project was conceived by by R.F. and developed by L.I., L.L., and R.F. The detailed simulations and calculations were carried out by L.I. and L.L. R.F. contributed to theory analysis of nonclassical features. The manuscript was written by L.I., L.L., and R.F.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41534-022-00538-y.

Correspondence and requests for materials should be addressed to Luca Innocenti.

Reprints and permission information is available at http://www.nature.com/reprints

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.