# Nonclassicality of Quantum States: A Hierarchy of Observable Conditions

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A necessary and sufficient hierarchy of conditions is derived that is completely equivalent to the failure of the Glauber-Sudarshan $P$ function to be a probability density. The conditions are formulated in terms of experimentally accessible characteristic functions of quadratures.

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The question of how to distinguish quantum states of a quantized harmonic oscillator, such as a mode of radiation or of atomic motion in a trap, having a classical counterpart from those displaying nonclassical properties has been a central issue in quantum optics over many years [1–21]. On one hand, rather general criteria were proposed [1–8], whose experimental verification may be cumbersome. On the other hand, observable criteria were formulated [9–15], which are based on specific observables. Particular nonclassical phenomena have been measured, such as photon antibunching [9], sub-Poissonian photon statistics [10], quadrature squeezing [11], negative values of the Wigner function [13], and photon number oscillations [16]. These features, however, only visualize specific aspects of nonclassicality and do not characterize the general properties of the quantum state.

A broadly accepted definition of nonclassicality of a quantum state relies on the Glauber-Sudarshan $P$ function [22]. An arbitrary density operator $\hat{\varrho}$ describing a single oscillator mode can be given in the representation

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

which is diagonal in the coherent states $|\alpha\rangle$. The real-valued and normalized function $P(\alpha)$ contains the complete information on the quantum state. It can be used to express any expectation value of a normally ordered operator function, $:\hat{f}(\hat{a}, \hat{a}^\dagger):$, of the annihilation and creation operator $\hat{a}$ and $\hat{a}^\dagger$, respectively, in a form that formally corresponds to a classical mean value:

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

In general, however, the $P$ function may be more singular than a $\delta$ function and can attain negative values. In such cases, it fails to be a probability density. If $P(\alpha)$ is a probability density, then Eq. (2) implies a close correspondence between expectation values in quantum and classical physics.

Consequently, a quantum state is considered to be nonclassical if it cannot be written as a statistical mixture of coherent states [1–3], i.e., if the $P$ function does not show the properties of a classical probability density, $P_{\rm cl}(\alpha)$:

$$P(\alpha) \neq P_{\rm cl}(\alpha). \tag{3}$$

We emphasize, however, that beside this condition there is a second signature of nonclassicality. It has been stated by Mandel that any light field of a small photon number displays nonclassical features [2]. Instead, one may say that a quantum state is nonclassical if the ground-state (or vacuum) noise plays a significant role for describing its properties [17]. This formulation is directly related to the requirement of normally ordering for expressing mean values according to Eq. (2).

In the following, we will not further deal with this second signature, but we intend to reformulate the condition (3) in a form that is accessible to experiments. One of the main difficulties consists in the fact that, due to its highly singular behavior, in general $P(\alpha)$ cannot be reconstructed from measured quantities. Thus, it is useful to formulate criteria for nonclassicality on the basis of a set of observables, that completely characterizes the quantum state under study. A prominent example of such a set of observables are the phase-sensitive quadrature operators,

$$\hat{x}(\varphi) = \hat{a}e^{i\varphi} + \hat{a}^\dagger e^{-i\varphi}. \tag{4}$$

Knowledge of their probability distributions, $p(x, \varphi)$, for all values of the phase $\varphi$ in an interval of size $\pi$ is equivalent to the complete knowledge of the quantum state. Moreover, the quadrature statistics can be measured for radiation modes, atomic motion in a trap, and for other related systems (for a review, see [23]).

In a recent Letter [17], one of us made the attempt to formulate an observable criterion for nonclassicality in terms of the characteristic function $G(k, \varphi)$,

$$G(k, \varphi) = \langle e^{ik\hat{x}(\varphi)}\rangle, \tag{5}$$

of the quadratures. It has been found that the $P$ function cannot be interpreted as a probability density if there exist values of $k$ and $\varphi$ for which

$$|G(k, \varphi)| > G_{\rm gr}(k), \tag{6}$$

where

$$G_{\rm gr}(k) = e^{-k^2/2} \tag{7}$$

is the characteristic function of the ground (or vacuum) state. The criterion (6) is sufficient to describe the nonclassicality of important classes of quantum states, such as Fock states, quadrature squeezed states, coherent superpositions of coherent states, Gaussian mixed states, and others. Its relevance for the interpretation of measured data has been demonstrated very recently [18]. In particular, a statistical mixture of a single-photon state and the vacuum state behaves nonclassically according to this condition, even when the Wigner function is non-negative. However, condition (6) is not equivalent to the condition (3), since there exist quantum states [24,25] that violate the former condition but that are nonclassical according to the latter. Thus, the problem of completely characterizing the failure of the $P$ function to be a probability density in terms of measurable quantities has not been solved yet.

The aim of this contribution is to present a solution of this fundamental problem. Our main result consists in a hierarchy of conditions for nonclassicality, formulated in terms of observable characteristic functions of quadratures. The hierarchy is completely equivalent to the condition (3); in the lowest order it reproduces the criterion (6) introduced in [17]. It characterizes nonclassicality of a quantum state on the basis of the same set of measured quantities already used in [18]. We illustrate the efficiency of our approach for the only example of a nonclassical state published yet [24] that violates the criterion (6).

Let us start with a reformulation of the nonclassicality condition (3) for the function $P(\alpha) \equiv P(\alpha_{\rm r}, \alpha_{\rm i})$. For this purpose, we introduce its characteristic function,

$$\Phi(u, v) = \int_{-\infty}^{\infty} P(\alpha_{\rm r}, \alpha_{\rm i}) \exp[2i(v\alpha_{\rm r} - u\alpha_{\rm i})] d\alpha_{\rm r} d\alpha_{\rm i}, \tag{8}$$

i.e., its twofold Fourier transform. It obeys the conditions

$$\Phi(0, 0) = \mathrm{Tr}\{\hat{\varrho}\} = 1, \qquad \Phi(-u, -v) = \Phi^*(u, v). \tag{9}$$

Now we express the nonclassicality condition (3) in terms of the characteristic function $\Phi(u, v)$. A theorem by Bochner [26] states that a continuous function $\Phi(u, v)$, obeying the condition $\Phi(0, 0) = 1$, is a classical characteristic function (and thus the Fourier transform of a probability density) if and only if it is positive semidefinite. This requires that, for arbitrary real numbers $u_k$ and $v_k$, arbitrary complex numbers $\xi_k$ $(k = 1, ..., n)$, and for any integer $n$, the condition

$$\sum_{i,j=1}^{n} \Phi(u_i - u_j, v_i - v_j)\, \xi_i\, \xi_j^* \geq 0 \tag{10}$$

is fulfilled. In other words, $P(\alpha_{\rm r}, \alpha_{\rm i})$ has all the properties of a probability density if and only if $\Phi(u, v)$ is positive semidefinite. Then the state under study is said to have a classical analog. Vice versa, if $\Phi(u, v)$ fails to be positive semidefinite, then $P(\alpha)$ is not a probability density.

If we associate with $\Phi(u, v)$ an $n \times n$ matrix with elements $\Phi_{ij} = \Phi(u_i - u_j, v_i - v_j)$, the condition (10) in more compact form reads as

$$\sum_{i,j=1}^{n} \Phi_{ij} \xi_i \xi_j^* \geq 0. \tag{11}$$

The left-hand side is a Hermitian form, $\Phi_{ji} = \Phi_{ij}^*$, cf. Equation (9). Now we can use the following theorem (e.g., see [27]): An $n \times n$ complex matrix is positive semidefinite if and only if the determinant of every of its principal submatrices is non-negative. In particular, this implies that the relation (11) is fulfilled, if and only if for any order $k = 2, \ldots, n$ the conditions

$$D_k = \begin{vmatrix} 1 & \Phi_{12} & \cdots & \Phi_{1k} \\ \Phi_{12}^* & 1 & \cdots & \Phi_{2k} \\ \cdots & \cdots & \cdots & \cdots \\ \Phi_{1k}^* & \Phi_{2k}^* & \cdots & 1 \end{vmatrix} \geq 0 \tag{12}$$

are valid. Note that the case $k = 1$ is irrelevant, since it leads to $D_1 = 1$, independent of the quantum state.

Thus, we may reformulate the necessary and sufficient condition for the classicality of a quantum state. A quantum state is classical, i.e., its $P$ function is a probability density, if and only if the condition (12) is fulfilled for all values of $k = 2, \ldots, \infty$. Based on this result, we arrive at the following necessary and sufficient criterion for nonclassicality. A quantum state is nonclassical if and only if there exist values $u_i$, $v_i$ $(i = 1, \ldots, k)$ for which at least one of the determinants $D_k$ $(k = 2, \ldots, \infty)$ becomes negative:

$$D_k < 0. \tag{13}$$

We may define nonclassicality of order $k - 1$ $(k = 2, \ldots, \infty)$ just by the condition (13). The higher the order of nonclassicality (and thus of the determinant), the more points of the characteristic function are included in the corresponding condition and the finer details of the characteristic function are relevant. Thus, in fact we arrive at a hierarchy of conditions (13) for nonclassicality [28].

Next, we consider the conditions for nonclassicality of first and second-order more explicitly. Setting $u_1 - u_2 = u$ and $v_1 - v_2 = v$, the condition of first-order nonclassicality, $D_2 < 0$, simplifies to

$$|\Phi(u, v)| > 1. \tag{14}$$

The second-order condition, $D_3 < 0$, reads as

$$|\Phi(u_1, v_1)|^2 + |\Phi(u_2, v_2)|^2 + |\Phi(u_1 + u_2, v_1 + v_2)|^2 - 2\,\mathrm{Re}\{\Phi(u_1, v_1)\Phi(u_2, v_2)\Phi^*(u_1 + u_2, v_1 + v_2)\} > 1. \tag{15}$$

Here we have changed the notation $u_1 - u_2 \rightarrow u_1$ and $u_2 - u_3 \rightarrow u_2$ and analogously for $v$. Clearly, this condition could be further simplified by choosing special relations between the points in the $(u, v)$ plane. Then, however, one may partly lose information on the nonclassical effects of the considered order.

For simplicity, let us consider in the following the situation for quantum states that show rotational symmetry in phase space. We introduce the real variables $k$, $\varphi$ [cf. Equation (5)] via $u = k\sin\varphi$, $v = k\cos\varphi$. For $\varphi$-insensitive quantum states, we get $\Phi(u, v) = \Phi(k, 0) \equiv \Phi(k)$, with $\Phi(k) = \Phi(-k)$. Thus, the first-order nonclassicality condition (14) reads as

$$\tilde{N}_1(k) \equiv |\Phi(k)| > 1. \tag{16}$$

To simplify the second-order condition, we choose $u_1 = u_2 = u/2$ and $v_1 = v_2 = v/2$, so that the inequality (15) can be rewritten as

$$[1 - \Phi(k)][1 - 2\Phi^2(k/2) + \Phi(k)] < 0. \tag{17}$$

Interestingly, in this case the determinant $D_3$ factorizes, where the first factor is closely related to the first-order condition (16). In particular, if the state fails to be nonclassical of first order $[|\Phi(k)| < 1]$, the factor $[1 - \Phi(k)]$ cannot be negative. In this case, second-order nonclassicality can arise only from the second factor. The condition reduces to

$$\tilde{N}_2(k) \equiv 2\Phi^2(k/2) - \Phi(k) > 1, \tag{18}$$

for our specific choice of points.

For example, let us consider the mixed state proposed by Diosi [24],

$$\hat{\varrho} = \sum_{n=1}^{\infty} 2^{-n}|n\rangle\langle n|, \tag{19}$$

representing a thermal state of mean excitation equal to one, whose ground (or vacuum) state has been suppressed via a measurement. The $P$ function of this rotationally symmetric state reads as

$$P(\alpha) = \frac{2}{\pi}e^{-|\alpha|^2} - \delta(\alpha). \tag{20}$$

Obviously, it describes a nonclassical state, since it fulfills the condition (3). The corresponding normally ordered characteristic function $\Phi(k)$ is given by

$$\Phi(k) = 2e^{-k^2} - 1. \tag{21}$$

It is easy to see that $|\Phi(k)| \leq 1$, so that the state (19) fails to obey the nonclassicality condition of first order.

Let us consider the properties of the same state with respect to nonclassicality of second order. From Fig. 1, it is seen that the inequality (18) is fulfilled over two semi-infinite intervals. Analytically, we get from Eq. (21) that the limit $k \to \pm\infty$ yields $\Phi(k) \to -1$ and thus $\tilde{N}_2(k) \to 3$. Therefore the state (19) clearly obeys the condition (18) for second-order nonclassicality.

Thus far, we have found an infinite hierarchy of conditions that a quantum state exhibits nonclassicality, formulated in terms of the normally ordered characteristic function $\Phi(u, v)$ of the $P$ function. In the next step we reformulate these conditions in terms of routinely measured quadratures. To be more specific, we will introduce the characteristic functions $G(k, \varphi)$ defined in Eq. (5). It is well known (for a review, see [23]) that this function is the product of the characteristic function $\Phi(k\sin\varphi, k\cos\varphi)$ and the (phase-independent) characteristic function $G_{\mathrm{gr}}(k)$ of the quadratures in the ground (or vacuum) state,

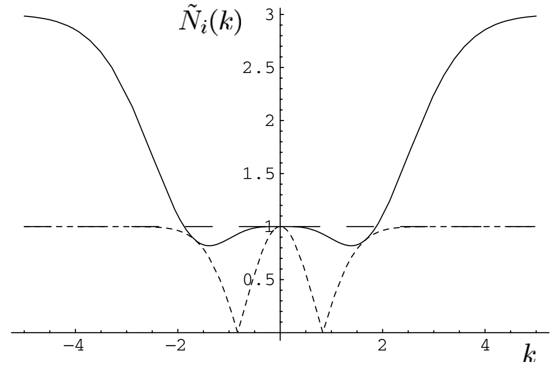


FIG. 1. First- and second-order nonclassicality conditions for the state (19). The dotted line shows $\tilde{N}_1(k)$, which never exceeds the classical limit of 1 (dashed line). The solid line displays $\tilde{N}_2(k)$, which exceeds the classical boundary of 1 over two semi-infinite intervals.

$$G(k, \varphi) = \Phi(k\sin\varphi, k\cos\varphi)\, G_{\mathrm{gr}}(k). \tag{22}$$

The characteristic functions $G(k, \varphi)$ of a freely propagating radiation mode can be derived from the data measured by balanced homodyne detection [18]. Moreover, these functions can even be directly measured for a cavity-field mode [29] or the quantized center-of-mass motion of a trapped ion in a harmonic potential [30].

Now we are able to express the first- and second-order condition for nonclassicality directly in terms of the measured characteristic functions $G(k, \varphi)$. Combining the first-order condition (14) with Eq. (22), we immediately find

$$N_1(k) \equiv |G(k, \varphi)| > G_{\mathrm{gr}}(k). \tag{23}$$

This exactly reproduces the nonclassicality condition (6), introduced in [17] and applied in experiments [18]. It states that a quantum state is nonclassical, if there exist values of $k$ and $\varphi$ for which the absolute value of the characteristic function exceeds the corresponding value in the ground (or vacuum) state.

The second-order condition (17), for rotationally symmetric states, in terms of the observable functions $G(k)$ reads as

$$[G_{\mathrm{gr}}(k) - G(k)] \times$$
$$[G_{\mathrm{gr}}(k) - 2G^2(k/2)G_{\mathrm{gr}}(k/\sqrt{2}) + G(k)] < 0, \tag{24}$$

where we have used the explicit form (7) of $G_{\mathrm{gr}}(k)$. In

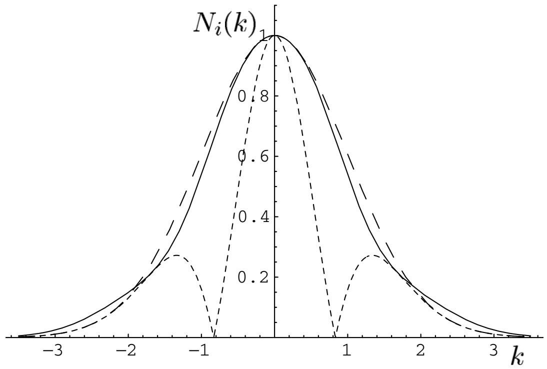


FIG. 2. Observable conditions for first- and second-order nonclassicality for the state (19). The dotted line represents $N_1(k)$, which never exceeds the classical limit $G_{gr}(k)$ (dashed line). The solid line displays $N_2(k)$, which exceeds $G_{gr}(k)$ within two semi-infinite $k$ intervals.

cases when the state under study does not display first-order nonclassicality, as for the example of the state (19), this condition is further simplified. On using Eq. (22), the condition (18) can be expressed as

$$N_2(k) \equiv 2G^2(k/2)G_{gr}(k/\sqrt{2}) - G(k) > G_{gr}(k), \qquad (25)$$

solely in terms of characteristic functions accessible to measurements. In principle, the same is also possible for any higher-order condition.

In Fig. 2, we show the quantities $N_1(k)$ and $N_2(k)$ for the state (19). Since $N_1(k) \equiv |G(k)| \leq G_{gr}(k)$, this state exhibits no first-order nonclassicality. However, $N_2(k)$ clearly exceeds the value $G_{gr}(k)$ over two semi-infinite intervals of $k$ values. This demonstrates second-order nonclassicality of the state, which is a clear signature for the existence of a nonclassical $P$ function, cf. Equation (20). The effect seems to be quite small, but we stress that even smaller effects of first-order nonclassicality have been clearly observed [18].

In conclusion, we have derived a hierarchy of observable conditions for a quantum state to be nonclassical. These conditions allow one to verify whether or not the Glauber-Sudarshan $P$ function is a probability density. The hierarchy of conditions naturally yields a classification of nonclassicality with respect to first, second, and higher orders. The method is illustrated for the example of a mixed state, which is classical in the first but nonclassical in the second order.

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