

Nonclassical character of statistical mixtures of the single-photon and vacuum optical states

A. I. Lvovsky*

Fachbereich Physik, Universität Konstanz, D-78457 Konstanz, Germany

J. H. Shapiro

Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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We demonstrate, theoretically and experimentally, that statistical mixtures of the vacuum state $|0\rangle$ and the single-photon Fock state $|1\rangle$ are nonclassical according to the Vogel criterion [W. Vogel, *Phys. Rev. Lett.* **84**, 1849 (2000)], regardless of the vacuum fraction. The ensembles are synthesized via conditional measurements on biphotons generated by means of parametric down conversion, and their quadrature statistics are measured using balanced homodyne detection. A comparative review of various quantum state nonclassicality criteria is presented.

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I. INTRODUCTION

Nonclassical states of the electromagnetic field have for many years provided an excellent playground for testing fundamental concepts of quantum mechanics [1]. Recently, nonclassical light has also become an important asset to the rapidly developing applied fields of quantum optics, such as quantum communication and quantum information technology. Its applications, to name only a few, include quantum cryptography [2], interferometric measurements [3], and linear-optics quantum computation [4].

Upon this background, it is becoming more important to have simple criteria for identifying a particular quantum state as nonclassical. This question has attracted strong interest from the first days of quantum optics and a number of solutions, both for the single-mode and multimode cases, have been proposed [6–11]. Although there exists a commonly accepted formal definition of a nonclassical state, no necessary and sufficient criterion has been proposed that would allow verification of an optical state's nonclassical character in a simple experiment.

For single-mode optical fields, nonclassical states are commonly defined as those that cannot be represented as a statistical mixture of coherent states. This can be reformulated in terms of the Glauber-Sudarshan P function [5]: if the latter is positive definite, i.e., if it can be interpreted as a probability density, then the state possesses a classical analog.

The practical application of the above definition requires complete information about the quantum state in question so that the P function can be reconstructed. In experimental practice, however, only partial information about a quantum state is normally available and it is often degraded by detection noise and losses. The goal of a nonclassicality criterion is to enable a conclusion about the character of a quantum state based only on this partial information. Known signatures of nonclassicality, frequently associated with a particular method of quantum state characterization, include anti-

bunching and sub-Poissonian photon statistics [6,7], squeezing [8], photon number oscillations [9], and negative values of the Wigner function [10,12].

All of these nonclassicality conditions are sufficient but not necessary and leave wide classes of quantum states outside their scope. For example, the Fock states are sub-Poissonian, but exhibit quadrature noises above the shot-noise level; phase-squeezed states are nonclassical, but possess super-Poissonian photon statistics and positive Wigner functions. This situation was recently improved by Vogel, who showed that a quantum state is nonclassical if the absolute magnitude of the state's characteristic function exceeds that of the vacuum state at any point in inverse phase space [13]. This modification of the traditional squeezing criterion covers a very wide set of quantum optical states. As noticed by Diósi, however, there are quantum states that are nonclassical but do not satisfy the Vogel criterion, so the latter is still not a necessary condition for nonclassicality [14].

The Vogel criterion is now very relevant because of recent progress in quantum homodyne tomography of highly nonclassical optical states. In a recent experiment, Lvovsky and coworkers prepared the single-photon Fock state $|1\rangle$ by conditional measurements on a photon pair produced via parametric down conversion [12]. The phase-averaged Wigner function reconstructed in the measurement showed a strong dip around the phase-space origin, reaching classically impossible negative values.

Although negativity of the Wigner function is a very strong indication of a quantum state's nonclassical character, it covers many fewer states than does the Vogel condition. As an example, we consider statistical mixtures of the single-photon Fock state $|1\rangle$ and the vacuum state $|0\rangle$

$$\hat{\rho}_{\text{mix}} = \eta|1\rangle\langle 1| + (1 - \eta)|0\rangle\langle 0|, \quad (1)$$

for $0 < \eta \leq 1$. The Wigner function associated with this mixed state takes on negative values only for $\eta > 0.5$ [15]. Moreover, reconstruction of the Wigner function requires acquisition of a set of phase-stabilized marginal distributions associated with different quadratures, or (as in [12]) the as-

*Email address: Alex.Lvovsky@uni-Konstanz.de

sumption that the Wigner function is rotationally invariant and hence deducible from a single phase-randomized marginal distribution. On the other hand, as we demonstrate in this paper, the Vogel criterion establishes the nonclassical character of ensembles (1) for any $0 < \eta \leq 1$, and it does so without requiring any assumptions regarding rotational symmetry or local-oscillator phase stabilization.

II. HOMODYNE DETECTION AND NONCLASSICALITY

Vogel's nonclassicality condition is applicable to quantum state data obtained from balanced homodyne detection (BHD). BHD is a technique for performing phase-sensitive measurements, i.e., field quadrature measurements, on a light wave. Proposed by Yuen and Chan in 1983 [16], it has become one of the main techniques for quantum state characterization. It has been used to detect squeezed states of the electromagnetic field [17], to characterize various quantum optical states tomographically [18], to demonstrate Einstein-Podolsky-Rosen-type quantum correlations [19], and to carry out continuous-variable quantum teleportation [20].

To perform BHD, the electromagnetic wave whose quantum state is to be determined is overlapped on a beam splitter with a relatively strong local oscillator (LO) wave in the same optical mode. The two fields emerging from the beam splitter are incident on two high-efficiency photodiodes whose output photocurrents are subtracted. The photocurrent difference is proportional to the value of the electric-field quadrature operator $\hat{E}(\theta)$ in the signal mode, where θ is the relative optical phase between the signal and LO. Repeated homodyne measurements on the same quantum state yield the statistical properties of the quantum noise associated with this quadrature. A set of quadrature noise measurements acquired at different values of θ is sufficient to reconstruct the Wigner function and the density matrix of the quantum state [21].

To demonstrate the nonclassical character of quantum states measured via BHD, comparison with the semiclassical theory of photodetection can be used. According to this theory, light is a classical electromagnetic wave and the detection noise arises from the discrete nature of the charge carriers created in the detection process. Assuming that BHD is carried out in the time-resolved, pulsed regime, the number of photoelectrons N_i produced by detection of the i th optical pulse consists of two independent parts: the quadrature measurement $N_{i,E}$ of the classical signal field plus the photoelectron shot-noise $\delta N_{i,S}$,

$$N_i = N_{i,E} + \delta N_{i,S}. \quad (2)$$

Assuming the signal field to be much weaker than the local oscillator, we can regard the shot noise as being entirely due to the LO. A local oscillator pulse of energy E_{LO} generates an average of $N_0/2 = \eta E_{LO}/2\hbar\omega$ charge carriers in each photodiode, where η is their quantum efficiency and ω is the optical frequency. These charge carriers are subject to Poisson statistics and carry a mean-square fluctuation of $\langle \delta^2(N_0/2) \rangle = N_0/2$. When the two photocurrents are subtracted, the sum of their independent shot noises leads to a

total mean-square fluctuation strength of $\langle \delta^2 N_S \rangle = N_0/2 + N_0/2 = N_0$ per pulse. According to the semiclassical theory, therefore, the total mean-square photodetection noise,

$$\langle \delta^2 N \rangle = \eta E_{LO}/\hbar\omega + \langle \delta^2 N_E \rangle, \quad (3)$$

always equals or exceeds the shot-noise level observed when the signal field is vacuum ($N_E \equiv 0$).

Whenever the signal field is coherent, i.e., carries no classical noise, the prediction (3) of the semiclassical calculation is identical to that of the "correct" theory, in which both the detector and the light are treated quantum mechanically. In keeping with standard practice in quantum optics, we define an optical state to be *classical* if it is a statistical mixture of coherent states. Classical states, therefore, have BHD photocurrent noise that can be correctly quantified using the above semiclassical theory. A squeezed state, on the other hand, is an example of a nonclassical state because its quadrature noise at some phase angles falls below the shot-noise level.

A more general approach to nonclassicality has been proposed by Vogel, whose treatment is rederived in the following. Let $\text{pr}(N)$ be the probability of detecting exactly N photoelectrons in the BHD subtraction output. In semiclassical photodetection theory, the optical quadrature noise of the signal field is statistically independent of the charge-carrier shot noise, so there exists separate probability distributions $\text{pr}_E(N)$ for the field noise and $\text{pr}_S(N)$ for the shot noise. According to Eq. (2), the measured $\text{pr}(N)$ is then the convolution of these two distributions

$$\text{pr}(N) = \text{pr}_E(N) \otimes \text{pr}_S(N). \quad (4)$$

Because $\text{pr}(N)$ and $\text{pr}_S(N)$ can be determined experimentally, the question of classicality reduces to the existence of a probability distribution $\text{pr}_E(N)$ that would satisfy the above equation.

Taking the Fourier transform of Eq. (4), we find

$$F[\text{pr}](\nu) = F[\text{pr}_E](\nu)F[\text{pr}_S](\nu), \quad (5)$$

where $F[\text{pr}]$ denotes the Fourier transform of the marginal distribution (i.e., the cross section of the state's Wigner function associated with the particular LO phase under investigation) and ν is the transform variable [22]. From probability-distribution normalization, which implies that $|F[\text{pr}_E](\nu)| \leq 1$ for all ν , we obtain a necessary condition for the classicality of a quantum state—the Vogel criterion [13]

$$|F[\text{pr}](\nu)| \leq |F[\text{pr}_S](\nu)| \quad (6)$$

for all ν . If inequality (6) is invalid for *any* quadrature at *any* value of ν , then Eq. (4) cannot be satisfied and the state is nonclassical.

An important feature of this nonclassicality condition (not mentioned in the original Vogel paper) is that it can be generalized to statistical mixtures of several marginal distributions associated with different phases. In other words, there is no need to maintain a stable phase relation between the LO and the optical state being tested. As soon as the histogram

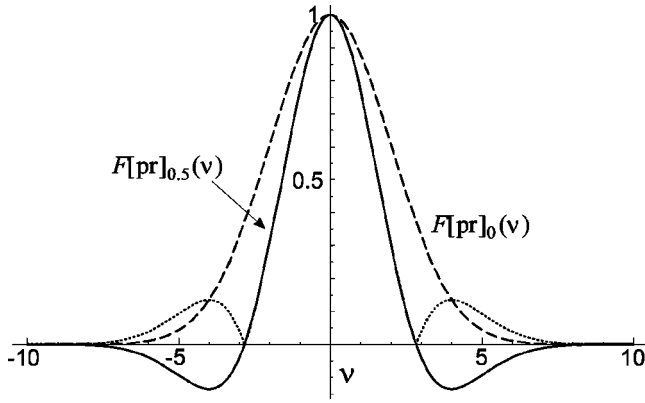


FIG. 1. Vogel nonclassicality of the state $\hat{\rho}_{\text{mix}} = \eta|1\rangle\langle 1| + (1 - \eta)|0\rangle\langle 0|$ with $\eta = 0.5$. Cross section of the characteristic function of this state (solid line) is shown along with that of the vacuum state (dashed line). The dotted line displays the absolute value of $F[\text{pr}]_{\eta}(\nu)$, which exceeds its vacuum state counterpart over two semi-infinite intervals.

of the quadrature measurements acquired in an experimental run violates inequality (6), the state is known to be nonclassical.

III. STATISTICAL MIXTURES OF VACUUM AND SINGLE-PHOTON STATES

A. Theoretical analysis

The ensembles defined by Eq. (1) are completely characterized by the Wigner function

$$W_{\eta}(X, P) = \frac{2}{\pi} (4\eta(X^2 + P^2) + 1 - 2\eta) e^{-2(X^2 + P^2)} \quad (7)$$

which is equivalent to the following, phase-independent marginal distribution for a quadrature measurement:

$$\text{pr}_{\eta}(X) = \sqrt{\frac{2}{\pi}} (1 - \eta + 4\eta X^2) e^{-2X^2}. \quad (8)$$

The mean-square quadrature deviation of the above distribution is

$$\langle \delta^2 X \rangle = 1/4 + \eta/2, \quad (9)$$

which always exceeds the vacuum-state value of 1/4. On the other hand, the Fourier image of this distribution,

$$F[\text{pr}]_{\eta}(\nu) = \int \text{pr}_{\eta}(X) e^{i\nu X} dX = (1 - \eta\nu^2/4) e^{-\nu^2/8}, \quad (10)$$

has an absolute value that exceeds that of the vacuum state ($F[\text{pr}]_0(\nu) = e^{-\nu^2/8}$) over two semi-infinite intervals of ν (Fig. 1). A simple calculation shows that for a given η , the difference between the characteristic functions $D_{\eta}(\nu)$

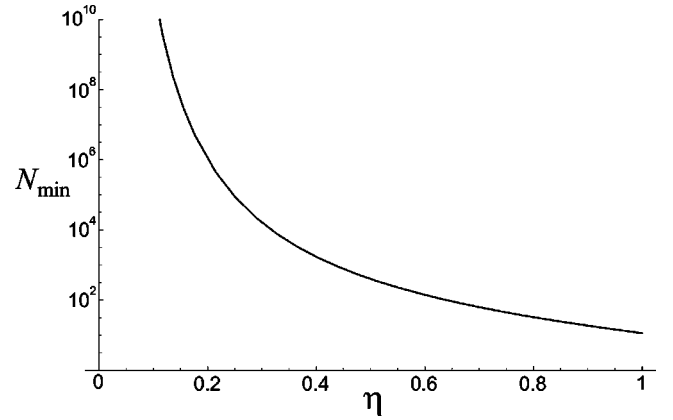


FIG. 2. Minimum number of quadrature samples that must be acquired in order to establish the nonclassical character of the state (1) beyond the margin of statistical uncertainty.

$= |F[\text{pr}]_{\eta}(\nu)| - |F[\text{pr}]_0(\nu)|$ reaches its maximum value of $D_{\eta}(\nu_{\text{opt}}) = 2\eta \exp[-(1 + \eta)/\eta]$ at

$$\nu_{\text{opt}}^2 = 8(1 + \eta)/\eta. \quad (11)$$

For any $0 < \eta \leq 1$, the quantity $D_{\eta}(\nu_{\text{opt}})$ is positive. Ensembles (1) are thus always nonclassical according to the Vogel criterion, even though they are never squeezed and their Wigner distribution is non-negative for $0 \leq \eta < 1/2$.

Experimentally, the characteristic function cross section is evaluated from a set of N individual quadrature measurements X_i as an estimate (empirical average)

$$\tilde{F}[\text{pr}]_{\eta}(\nu) = \langle e^{i\nu X_i} \rangle_N. \quad (12)$$

This evaluation suffers a mean-square estimation error

$$\langle |\tilde{F}[\text{pr}]_{\eta}(\nu) - F[\text{pr}]_{\eta}(\nu)|^2 \rangle = \frac{1}{N} (1 - |F[\text{pr}]_{\eta}(\nu)|^2). \quad (13)$$

The nonclassical character of a quantum state can be established only if this uncertainty does not exceed $D_{\eta}(\nu_{\text{opt}})$. Since the latter function decays quickly with reducing η , demonstrating the nonclassicality of ensembles (1) at low efficiencies requires an exponentially large number of samples to be acquired (Fig. 2).

B. Experiment

The experimental apparatus employed in our paper was almost identical to that described in [12] (Fig. 3). The 790-nm, 1.6-ps output of a Spectra Physics Tsunami laser was frequency doubled in a single pass through a 3-mm LBO crystal and then passed on to a 3-mm BBO crystal for down conversion. The down converter operated in a type-I frequency degenerate, noncolinear configuration. A single-photon counter (EG&G SPCM-AQ-131) was placed in one of the emission channels—labeled trigger—to detect photon-pair creation events and to trigger a homodyne system placed

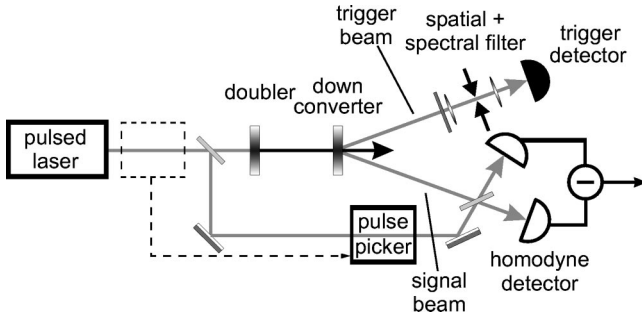


FIG. 3. Master scheme of the experimental setup. The position of the pulse picker allowed a much higher photon pair-production rate to be achieved.

in the other emission channel—labeled signal. In this way, pulses selected for homodyne measurements are only those for which a photon has been emitted into the signal channel, thus preparing single-photon Fock states by conditional measurements.

We used a small fraction of the original optical pulses from the pulse picker—split off before the frequency doubler—as the local oscillator for the homodyne system. These pulses have to be temporally and spatially mode matched to the mode of the photons in the signal channel [23]. In order to resolve the quantum noise of individual laser pulses, a time-domain homodyne detector with ultra-low electronic noise (~ 1000 electrons per pulse), high subtraction efficiency (>83 dB), and high-frequency bandwidth (DC–2 MHz) has been used [24].

The apparatus employed in [12] suffered a significant drawback owing to its very low pair-production rate (around one pair per four seconds). This was caused, in particular, by a deliberate factor-of-100 reduction of the Ti:sapphire master laser’s 82-MHz pulse repetition rate, which was achieved by using an acousto-optical pulse picker. The necessity for this reduction arose from the relatively low (~ 1 MHz) bandwidth of the balanced homodyne detector amplifier; the detector was unable to resolve the shot noise of individual laser pulses arriving at a higher rate.

In the present paper, the pair generation rate was dramatically increased by installing an electro-optical pulse picker in the beam path of the local oscillator, instead of right after the master laser. The entire setup, apart from the homodyne detector, was thus operating at the original laser repetition rate of 82 MHz. Whenever a trigger photon detection event occurred, the optical shutter in the local oscillator opened to transmit a single local oscillator pulse that activated a balanced homodyne measurement. In order to compensate for the trigger delay of the optical shutter, a 50-ns optical delay line was introduced. The replacement of the pulse picker and a number of other modifications allowed us to enhance the pair-production rate by a factor of more than a 1000 in comparison to the work [12]; the original experiment that lasted 14 hours would only last 45 seconds with the new apparatus.

Various experimental imperfections cause an admixture of the vacuum $|0\rangle$ to the Fock state $|1\rangle$ that would be measured if the setup were ideal [12]. When optimally aligned, our apparatus constructed ensembles $\hat{\rho}_{\text{mix}}$ with the highest over-

all efficiency $\eta=0.61$. Further reductions of η —to probe the Vogel criterion for $\hat{\rho}_{\text{mix}}$ at high-vacuum fraction—have been achieved by misaligning the temporal synchronization between the LO and single-photon pulses.

We acquired nine data sets, each containing 100 000 electric-field quadrature measurements for the states (1) with the measurement efficiencies η between 0.19 and 0.61 and a single data set of 200 000 points for the vacuum state. The phase-averaged Wigner functions reconstructed from the data sets corresponding to $\eta=0.58$ and 0.61, as expected, exhibited negativities around the phase-space origin point. All data sets were then binned up to obtain their histograms [Fig. 4(a)]. The Fourier images $|\tilde{F}[\text{pr}]_{\eta}(\nu)|$ of the marginal distributions, calculated using Eq. (12) and shown in Fig. 4(b), clearly violate inequality (6). The nonclassicality of the states investigated is made manifest in Fig. 4(c), where the values of $|\tilde{F}[\text{pr}]_{\eta}(\nu_{\text{opt}})|$, reflecting the highest difference between the classical and quantum behavior, are plotted.

Because the setup was not interferometrically stable, each of the quadrature data sets corresponds to the superposition of marginal distributions of ensembles (1) associated with different phases. This circumstance does not compromise our conclusion on the nonclassical character of these states, as the latter does not rely on any assumptions regarding phase stability or randomness.

Of special interest are the quadrature data acquired for the 0.28 and 0.19 efficiency values. The marginal distributions associated with these efficiencies [Fig. 4(a)] are not only wider than that of the vacuum state but also lack any fine structure which would allow a direct decision on their nonclassical character. Only the Fourier analysis of these distributions allows us to draw such a conclusion. These examples show that the suggested reformulation of Vogel’s criterion, stating that “a quantum state has no classical counterpart when these [marginal distribution] functions show structures that are narrower than the corresponding distributions of the ground state of the oscillator” [13] is not always rigorous.

IV. DISCUSSION

Comparing the Vogel criterion and the negativity of the Wigner function as nonclassicality conditions, we first notice that fulfillment of either one does not automatically imply satisfaction of the other. In the previous section we have considered quantum states that are nonclassical according to the Vogel criterion but have positive-definite Wigner functions. As an opposite example one can use the ensemble presented in the Diósi correspondence on the Vogel paper [14]

$$\hat{\rho} = \sum_{n=1}^{\infty} 2^{-n} |n\rangle\langle n|. \quad (14)$$

The Glauber-Sudarshan P function for this state is

$$P_{\text{GS}}(X, P) = \frac{2}{\pi} e^{-(X^2 + P^2)} - \delta(X)\delta(P). \quad (15)$$

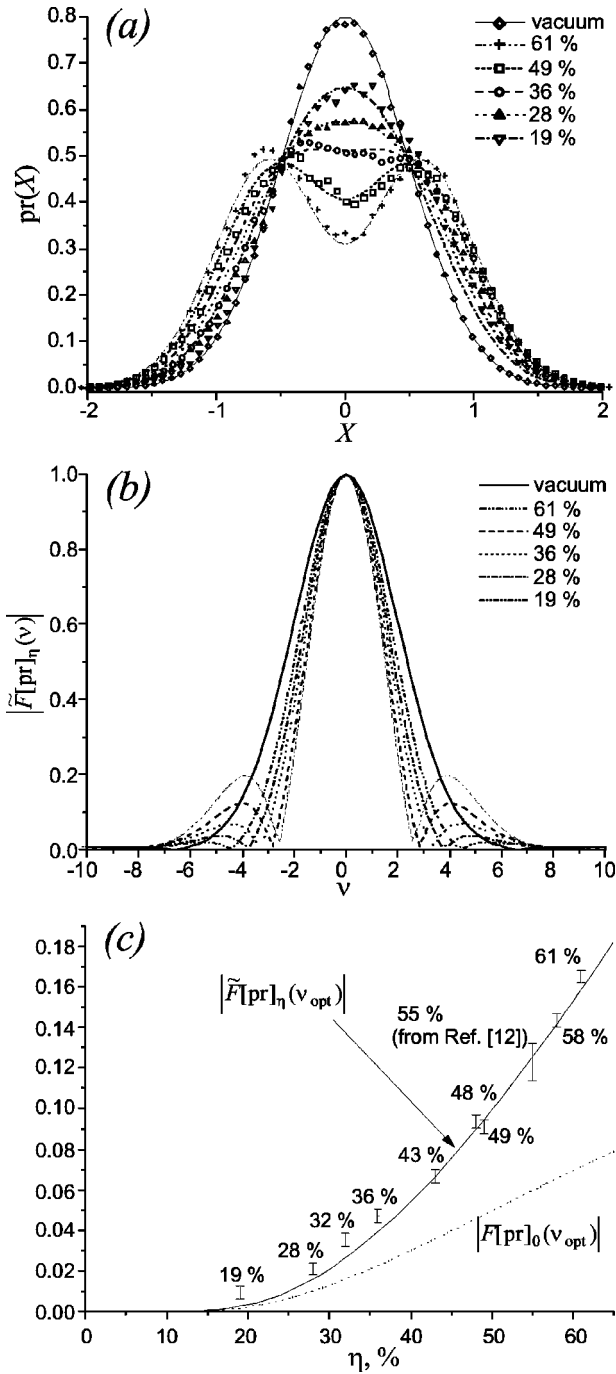


FIG. 4. Experimental results. (a) Marginal distributions associated with the vacuum state and ensembles (1) for five different efficiency values; (b) Cross sections of the characteristic functions obtained through Fourier analysis of the homodyne data; (c) Theoretical (solid line) and experimental values of $|\tilde{F}[\text{pr}]_\eta(v_{\text{opt}})|$. The function $|F[\text{pr}]_0(v_{\text{opt}})|$ (dashed line), which establishes the classical limit, is also displayed.

Convolving this P distribution with a circularly symmetric zero-mean variance $1/4$ two-dimensional Gaussian function gives the Wigner distribution [21]

$$W(X, P) = \frac{4}{3\pi} e^{-2(X^2 + P^2)/3} - \frac{2}{\pi} e^{-2(X^2 + P^2)}. \quad (16)$$

This distribution is negative at $X=P=0$, but (as shown by Diósi) the above state fails the Vogel criterion for nonclassicality.

It is also instructive to compare the two nonclassicality criteria from a purely classical viewpoint. To what extent can the fulfillment of each condition persuade someone who only believes in classical physics that something is wrong with her picture of the world? In this aspect, the two criteria are strikingly different in terms of fundamental assumptions the “classicalist” must maintain in order to be convinced of a contradiction by observing one or the other criterion fulfilled. The Vogel condition is based on the semiclassical theory of photodetection, i.e., it assumes that homodyne detection results in field-quadrature observation that is embedded in a certain, well-defined amount of (shot) noise. Homodyne tomography, from which the Wigner distribution in reconstructed, makes no such postulate, and can be fully interpreted in the framework of classical physics. In particular, this classical interpretation makes the Wigner distribution a (positive-definite) classical probability distribution over phase space. All that is presumed in making this classical interpretation are Maxwell’s equations for the electromagnetic wave and the fact that a photodiode’s output current is proportional to the intensity of the incident beam (perhaps with some added classical noise). The combination of a classically understandable measurement method and an evident non-classicality of its result—the negative Wigner function, such as in the work [12]—provides very strong evidence of quantum mechanics.

In the framework of this discussion it is interesting to compare the Wigner function’s negativity with another well-known signature of nonclassicality, namely, the violation of Bell’s inequality. In the latter, the assumptions that are made are even less restrictive. Causality is the only physical postulate; no assumptions need to be made regarding the physical nature of the experiment itself. If a tomographic reconstruction of a Wigner function with negative values would convince a *physicist* that the world is quantum, a loophole-free violation of the Bell inequality would convince a *philosopher* who has no knowledge of physics whatsoever. From a purely philosophical point of view, tomographic reconstruction of a negative Wigner function is stronger evidence of quantum mechanics than is the incompatibility of the detection noise distribution with the semiclassical photodetection theory, but weaker than violation of Bell’s inequalities.

In terms of experimental simplicity, however, the sequence goes the other way. Apart from the fact that quantum states with negative Wigner functions (e.g., Fock states) are quite exotic and difficult to synthesize, the complete reconstruction of the Wigner distribution requires acquisition of a full set of marginal distributions with fixed relative phase between the LO and the state being examined. On the other hand, verification of the Vogel criterion can be applied to a single marginal distribution or to a superposition of marginals associated with different phases. To date, no loophole-free violation of Bell’s inequalities has been reported [25].

V. CONCLUSIONS

We have shown that the electric-field-quadrature statistics of the ensembles (1) of the vacuum state $|0\rangle$ and the single-photon Fock state $|1\rangle$ are nonclassical according to the Vogel criterion [13]. These states were synthesized and measured in a setting similar to that of Lvovsky *et al.* [12] but with an improved pair-production rate achieved thanks to a method of pulse picking. The Vogel criterion has been generalized to apply to quadrature distributions obtained in interferometri-

cally unstable settings. A quantitative analysis of statistical errors has been given.

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