Highly nonclassical photon statistics in parametric down-conversion

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We use photon counters to obtain the joint photon-counting statistics from twin-beam nondegenerate parametric down-conversion, and we demonstrate directly, and with no auxiliary assumptions, that these twin beams are nonclassical.

I. INTRODUCTION

The quantum nature of light is exemplified by the fact that it can exhibit both undular (wavelike) and corpuscular (particlelike) properties under different circumstances. Using an atomic cascade to produce a pair of nearly simultaneous photons [1], Grangier *et al.* [2] demonstrated that a heralded photon can either interfere with itself in a Mach-Zehnder interferometer or be shown to either wholly transmit or reflect by a beam splitter, which demonstrates complementarity for light. More recently the continuous trade-off of undular and corpuscular features in a quantum nondemolition measurement [3] was demonstrated using a nondeterministic linear optical gate [4]. Technically, nonclassical light corresponds to field states whose Glauber-Sudarshan *P* representation is singular [5,6]; practically, nonclassicality can be established by demonstrating photon antibunching [7] or using optical homodyne tomography [8,9] to demonstrate that the Wigner function (which is a Gaussian convolution of the *P* representation) exhibits squeezing [10] or is negative over some region [11].

Recently Waks, Diamanti, Sanders, Bartlett, and Yamamoto (WDSBY) [12] exploited cutting-edge photon-counting technology [using the visible light photon counter (VLPC) [13]] to demonstrate that a single beam emitted by parametric down-conversion (PDC) is nonclassical by measuring its photon statistics and avoiding auxiliary assumptions such as detector efficiency. Here we reinterpret the WDSBY data using Klyshko's powerful criterion for direct detection of nonclassicality via photon statistics [14], and we demonstrate nonclassicality of twin-beam PDC by using two photon counters and Lee's extension of Klyshko's criterion to two beams [15].

Although the simultaneity of photon pair production by twin-beam, or nondegenerate, PDC is well known [16], leading to its applications as a heralded photon source [17] and for tests of Bell's inequalities [18], the joint photon statistics of the twin beams have not previously been directly measured. These statistics are necessary for a direct confirmation of the nonclassicality of twin-beam PDC, which is one of the most important tools in quantum optics as a source of correlated or entangled pairs of photons [19] and, more recently, as a source of entangled fourtuples of photons [20]. We have performed this direct photon-counting experiment and obtained joint photon-counting data for the twin beams and thereby showed that these twin beams are truly nonclassical.

II. SINGLE-BEAM NONCLASSICAL STATISTICS

Theories generally treat the twin-beam PDC output as a two-mode squeezed vacuum state [21]

$$|\eta\rangle = \sqrt{1 - |\eta|^2} \sum_{k=0}^{\infty} \eta^k |kk\rangle, \qquad (1)$$

with $\eta \in \mathbb{C}$ and the argument of η dependent on the phase of the pump laser field for the PDC. For degenerate PDC, the state (1) is replaced by

$$|\chi\rangle = \sqrt{1 - |\chi|^2} \sum_{k=0}^{\infty} \chi^k |2k\rangle, \qquad (2)$$

and the photon number distribution is

$$p_n = (1 - |\chi|^2)|\chi|^{2n}. \qquad (3)$$

This state corresponds to a joint photon number distribution for the two beams, which is given by

$$p_{n_1, n_2} = (1 - |\eta|^2)|\eta|^{2n_1} \delta_{n_1, n_2}. \qquad (4)$$

Klyshko considered a state of the type emitted by an ideal degenerate PDC, and Lee's analysis of photon-counting correlations explicitly assumed two-mode states and included a treatment of the two-mode squeezed vacuum (1).

The reality of PDC and photon counting is somewhat more complicated. The twin beams (known as signal and idler fields) are each multimode so the theoretical treatments of these outputs are not immediately applicable. In fact there are so many modes for each of the signal and idler beams that we observe a photon distribution over these modes that is Poissonian. The Poisson distribution originates from the fact that there so many signal and idler modes that the average number of pairs per mode is much less than 1. This means that each mode is predominantly a vacuum state with a small one-photon contribution and a negligible multiphoton contribution. Thus, the generated signal is created by a sum of independent spontaneous emitters (one for each mode), and, as each pair is created independently, the pair creation statistics must be a Poisson distribution.

This discrepancy in photon statistics between the usual theoretical treatment of PDC output [21] and experimental reality makes the direct measurement of joint photon-counting statistics and verification of nonclassicality even more interesting and important. Properties of twin-beam

PDC have been explored in an elegant experiment by Haderka *et al.* [22], but their approach is different in that they do not use two distinct photon counters on each beam. Thus, inferences of nonclassicality rely on auxiliary assumptions that are not required by our approach such as taking into account the losses during transmission, quantum efficiency, and internal noise of the camera and noise due to other light sources to obtain the joint signal-idler photon number distribution [22].

To begin, we reanalyze the WDSBY data for single-beam PDC. WDSBY [12] conclusively demonstrated nonclassicality using their data for one-, two-, and three-photon measured counts $\{\wp_n; n=1,2,3\}$, where we use the symbol p for the ideal probability and $\wp$ for the measured probability. We will now compare their results to Klyshko's criterion [14]. We introduce Klyshko's criterion for two reasons: (i) Klyshko's criterion demonstrates that the WDSBY data violate classical bounds beyond the three-photon case studied by WDSBY, and (ii) Klyshko's criterion is a basis for Lee's criterion [15] which we use for the analysis of the twin beams.

WDSBY used the fact that nonclassicality of light, defined by the singularity of the Glauber-Sudarshan P representation, holds if and only if the photocount distribution for the beam cannot be expressed as a sum or integral of Poisson distributions (which corresponds to classical count distributions). Specifically the photon count statistics $\{\wp_n; n=1,2,3\}$ were irreconcilable with classical photon statistics; thus photon counting provided a direct means for establishing that a single-beam light source is nonclassical.

To demonstrate nonclassical photon statistics, WDSBY pumped a type-I phase-matched Beta Barium Borate (BBO) crystal set up for collinear degenerate amplification with 20 ns pulses of the fourth harmonic (266 nm) of a Q-switched neodymium-doped yttrium aluminum garnet (Nd:YAG) laser. In this configuration, the down-converted photons had half the energy of the pump (532 nm) and traveled in the same direction. The pump was removed by a prism, while the down-conversion was focused by a 250 mm lens onto the VLPC detector. The detector output was amplified and then sent to a gated boxcar integrator, which was triggered by the laser. This configuration was used to measure the photon number distribution of the detected field. WDSBY achieved a 40 standard deviation violation of classicality, according to the criterion

$$\Gamma = \frac{\wp_2}{\wp_1 + \wp_2 + \wp_3} > \Gamma_{\text{classical}} \equiv \frac{3}{3 + 2\sqrt{6}} \approx 0.379 \quad (5)$$

without any auxiliary assumptions about the detector or field.

In fact Klyshko's criterion preceded the WSBY criterion and was the first proposal for a direct test of nonclassicality according to "local properties" of the photon number distribution (PND) [14],

$$K_n = (n+1)\frac{p_{n-1}p_{n+1}}{np_n^2} < 1, \quad n = 1,2,3,\dots. \quad (6)$$

By the replacement $p \mapsto \wp$, the criterion applies to measured photon statistics rather than the ideal photon-counting distri-

bution. Lee [15] refers to such tests of nonclassicality according to local properties of the PND as type II, as opposed to the traditional version (which Lee calls type I) that employs inequalities on moments (such as the Mandel Q parameter [23]).

If criterion (6) is satisfied for any n, then the field is necessarily nonclassical. To compare with Γ, consider $n=2$ for Klyshko's criterion applied to measured data:

$$K_2 = \frac{3}{2}\frac{\wp_1\wp_3}{\wp_2^2} < 1. \quad (7)$$

Rearranging the terms in Eq. (7) yields the nonclassicality criterion

$$\wp_2 > \sqrt{\frac{3}{2}\wp_1\wp_3}. \quad (8)$$

An alternative criterion emerges from the nonclassicality criterion (5), which yields

$$\wp_2 > \frac{1}{2}\sqrt{\frac{3}{2}}(\wp_1 + \wp_3). \quad (9)$$

Thus, combining Klyshko's result for K_2 and the WDSBY result for Γ, we obtain the general condition, based on measured results $\{\wp_1, \wp_2, \wp_3\}$ for nonclassicality being

$$\wp_2 > \min\left\{\sqrt{\frac{3}{2}\wp_1\wp_3}, \frac{1}{2}\sqrt{\frac{3}{2}}(\wp_1 + \wp_3)\right\}. \quad (10)$$

This condition is based on the best case by combining the WDSBY Γ criterion (5) with Klyshko's criterion for $n=2$.

Equation (10) can be further simplified: the two quantities in this equation coincide in the symmetric case that $\wp_1 = \wp_3$; then $\wp_2 > \sqrt{3/2}\wp_1$. Now suppose that $\wp_3 = \kappa\wp_1$. We can thus rewrite condition (10) as

$$\frac{\wp_2}{\wp_2} > \frac{3}{2}\min\left\{\sqrt{\kappa}, \frac{\kappa+1}{2}\right\}. \quad (11)$$

Except for $\kappa = 0,1$ the first term on the left-hand side of Eq. (11) is always smaller than the second term, and the two terms are equal when $\kappa = 0,1$. Thus Klyshko's condition is stronger than the WDSBY criterion, and another advantage of Klyshko's general criterion over WDSBY's criterion is that the latter only applies in the measured photon number distribution localized around $n=2$ whereas Klyshko's criterion explores nonclassicality for local regions of the photon number distribution around any $n \geq 1$.

In Fig. 1, we present a plot of K_n vs n for the WDSBY data. Klyshko's criterion is violated for even numbers of photons between $n=2$ and 8. The limit of our measurement capability is nine photons, and all odd-number detections fail to violate Klyshko's criterion as expected from joint distribution (4). The reason why even-number counts violate the classical limit and odd-number counts do not is that the ideal photon number distribution is nonzero only for even-number counts. Experimentally we detect odd-number counts due to finite detector quantum efficiency, but this does not lead to violation of the classical limit. Figure 1 is especially important in demonstrating that nonclassicality of the beam is evi-
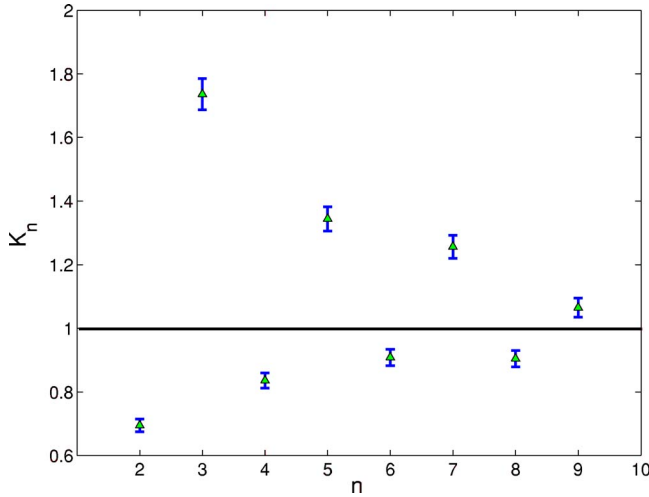
FIG. 1. (Color online) Experimental results for Klyshko's figure of merit K vs photon number n for a single-mode field.

dent for all even photon numbers up to the limits imposed by our photon counter capabilities.

III. TWIN-BEAM NONCLASSICAL STATISTICS

Now let us consider the twin-beam PDC and its joint photon-counting statistics. We will use Lee's generalization of Klyshko's type-II criterion to establish nonclassicality of the twin-beam PDC output directly from the joint photon-counting data with no adjustments made for photodetection efficiency. We will need to adapt Lee's criterion to accommodate the multimode aspect of the PDC output.

Lee introduces the nonclassical criterion for two modes of a light field given by

$$R_{n_1 n_2} = (n_2 + 1)\frac{p_{n_1-1,n_2+1}}{2n_1 p_{n_1,n_2}} + (n_1 + 1)\frac{p_{n_1+1,n_2-1}}{2n_2 p_{n_1,n_2}} < 1, \quad (12)$$

with $n_1, n_2 \in \{1, 2, 3, \ldots\}$. The field is nonclassical if $R_{n_1 n_2}$ satisfies inequality (12) for any n_1 and n_2. Although Lee's analysis supposes a two-mode field, this result is equally valid for a multimode field and the use of two photon counters, with some modes directed to one detector and other modes directed to the other detector.

Mode mismatches and lost modes are partially responsible for photon counter inefficiencies. As we do not employ auxiliary assumptions, the onus is on us to enhance efficiency to ensure that inequality (12) is satisfied, and not to resort to modeling these losses and adjusting the photon statistics according to these assumptions.

We define n_1 as the number of signal photons and n_2 as the number of idler photons. It is convenient to determine the conditional probabilities for n_2 photons given a count of n_1 at the other detector, denoted by $p_{n_2|n_1}$. The conditional and joint number-counting probabilities are related by the formula

$$p_{n_1,n_2} = p_{n_2|n_1} p_{n_1}. \quad (13)$$

As explained in the previous section, the counting statistics for each detector is a Poissonian distribution with
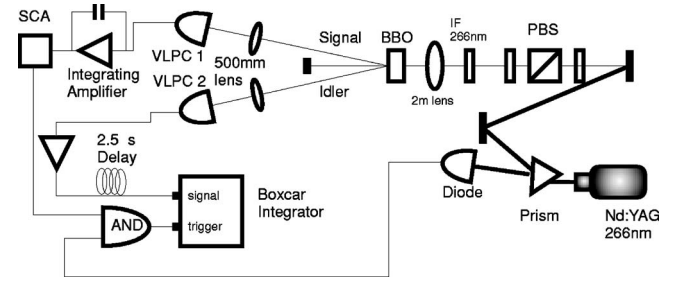

FIG. 2. Experimental setup for measuring violation of Lee's inequality.

$$\wp_n = \exp(-\bar{n})\bar{n}^n/n! \quad (14)$$

where $\bar{n}$ is the mean photon number summed over all the signal modes. The choice of the Poisson distribution is justified by the fact that there are many more signal and idler modes than photons distributed among these modes [22].

Lee's expression (12) can be revised in terms of conditional probabilities. Using the simple relation $\wp_n^{\text{Poisson}}/\wp_{n-1}^{\text{Poisson}} = \bar{n}/n$, we rewrite the criterion as

$$R_{n_1 n_2} = \frac{\bar{n}^2 \wp_{n_2-1|n_1+1} + n_1(n_1+1)\wp_{n_2+1|n_1-1}}{2\bar{n} n_2 \wp_{n_2|n_1}} < 1. \quad (15)$$

In our experiments, the mean photon number collected over all the modes is $\bar{n} = 1$.

The experimental setup for generating twin photon beams for measuring the Lee nonclassical criterion is shown in Fig. 2. A 266 nm pump source is generated from the fourth harmonic of a Q-switched Nd:YAG laser. The pump pulses have a duration of 20 ns and a repetition rate of 45 kHz. A dispersion prism separates the fourth harmonic from the residual second harmonic, which is used to illuminate a high-speed photodiode to generate a triggering signal. The fourth harmonic pumps a BBO crystal, which is set for noncollinear degenerate phase matching.

In this condition, the signal and idler waves are both 532 nm in wavelength and have a divergence angle of 1° from the pump. The pump is loosely focused before the BBO crystal to achieve a minimum waist at the collection lens. This results in a sharper two-photon image which enhances the collection efficiency. The pump power is set to 20 μW. Using the count rate on the detectors and known values of the detector quantum efficiency, the average pair creation rate at this pump power is measured to be one pair per pump laser pulse.

Two VLPC detectors are used in this experiment. Each detector is held in a separate helium bath cryostat and cooled down to 6–7 K, which is the optimum operating temperature. The VLPC is sensitive to photons with wavelengths of up to 30 μm, so it must be shielded from room-temperature thermal radiation. This is achieved by encasing the detector in a copper shield, which is cooled down to 6 K. Acrylic windows at the front of the copper shield are used as infrared filters. These windows are highly transparent at visible wavelengths and simultaneously nearly opaque at 2–30 μm wavelengths.

TABLE I. Experimental results for Lee's figure of merit R vs photon number n for twin beams.

n_1	n_2			
	1	2	3	4
2	0.69 ± 0.023	0.27 ± 0.007	0.47 ± 0.012	1.37 ± 0.04
3	2.23 ± 0.08	0.70 ± 0.022	0.33 ± 0.008	0.47 ± 0.012

VLPC 1 is used as the triggering detector which detects the number of photons generated in the signal arm on a given laser pulse. The output of VLPC 1 is amplified by an integrating amplifier, which generates an electrical pulse whose height is proportional to the number of emitted electrons. The height of the pulse is discriminated by a single-channel analyzer (SCA). A logical AND is performed between the output of the SCA and the output of the photodiode to reject all detection events which occur outside of the pulse duration.

The output of VLPC 2, which measures photons in the idler arm, is amplified and connected to the signal input of a boxcar integrator. On each pulse from the SCA, the output of VLPC 2 is integrated over a 2 μs window, which is sufficiently large to encompass the entire electrical pulse (determined by the bandwidth of subsequent amplifiers). The pulse area is proportional to the number of detected photons. By measuring the pulse area histogram of the VLPC, we can therefore measure $p_{n_2|n_1}$, where n_1 and n_2 are the number of photons in the signal and idler arms, respectively.

The experimental results are given in Table I. Since we can measure n_1 only from one to four photons, we obtain expressions for the Lee bound only for $n_1 = 2, 3$. In the ideal case where we have perfect detection efficiency and no dark counts, we would have $R_{n_1 n_2} = 0$ when $n_1 = n_2$. When $n_1 \neq n_2$ the Lee bound would not be well defined because $p_{n_1, n_2} = 0$, causing a divergence. In the presence of detection losses, we would still expect the best violations when $n_1 = n_2$, with worse results in the off-diagonal term. As can be seen in Table I, this is indeed the case. When $n_1 = n_2 \in \{2, 3\}$, we obtain extremely good violations of the Lee criterion, whereas off-diagonal terms yield worse violations or no violations at all.

V. CONCLUSIONS

In summary we have demonstrated directly that twin-beam PDC produces nonclassical light. As PDC is one of the most important tools for quantum optics, it seems surprising that the nonclassical nature of PDC output has not been directly verified before. One reason for not having previously established nonclassicality of twin-beam PDC is the requirement for sophisticated, modern photon counters and correlations of their data. Another reason is that only recently has direct testing of nonclassicality via the local properties of the measured photon count distribution been understood [12,14,15]. Our method of directly measuring photon counts for twin beams applies to multiple-beam fields and testing the local properties of the measured photon count distribution provides a valuable, practical means for establishing nonclassicality of light, especially in cases where the photons are not antibunched.

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