Direct Observation of Nonclassical Photon Statistics in Parametric Down-Conversion

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We employ a high quantum efficiency photon number counter to determine the photon number distribution of the output field from a parametric down-converter. The raw photocount data directly demonstrates that the source is nonclassical by 40 standard deviations, and correcting for the quantum efficiency yields a direct observation of oscillations in the photon number distribution.

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Quantum optics [1,2] is concerned with optical phenomena that cannot be described by a classical field treatment. Operationally, photon statistics are measured and nonclassicality is established by violations of inequalities imposed by the assumption that the field is classical. Well-known examples are observations of sub-Poissonian photon number statistics [3], photon antibunching [4], and photocurrent fluctuations below the shot noise level for squeezed light sources [5]. Although an equivalence between sub-Poissonian statistics and photon antibunching has sometimes been assumed, the two phenomena are distinct revelations of a nonclassical field [6,7], and the choice of test for nonclassicality may need to depend on the source. Both measures are adequate to test nonclassicality for resonance fluorescence, but antibunching is certainly not sufficient to identify nonclassicality for two-photon sources such as an atomic cascade [8] or parametric down-conversion (PDC) [9]. Sophisticated demonstrations of nonclassicality for two-photon sources have been employed such as showing complementarity [10] or violating a Bell inequality [11]. However, direct measurement of the photon number distribution may be sufficient to demonstrate nonclassicality, and such a measurement is appealing because the nonclassicality criterion is strictly based on the actual measurement record, inequalities that arise from a classical field treatment, and no assumptions whatsoever about the source, propagation, or prephotodetection processing.

Direct measurements of the photon number distribution have been elusive for technical reasons. But recent advances in photon counting technology, namely, the visible light photon counter (VLPC) [12,13], now provides a means for directly testing nonclassicality via photon counting for the (bunched) output field from a PDC, as we demonstrate here (direct testing is contrasted with indirect testing such as measures that rely on reconstruction of the density matrix [14]). Photon counting criteria can provide a sufficient but not necessary condition for demonstrating nonclassicality. Several criteria for proving nonclassicality of the source via direct counting are already known, such as sub-Poissonian statistics [15], observing photon number oscillations [16] and deducing if these oscillations are classical or nonclassical [17], or by employing Hillery’s two criteria of comparing the total probabilities of even vs odd photon numbers being detected [18]. However, such criteria are not necessarily practical, especially for a super-Poissonian two-photon source. We introduce a new and practical criterion for evaluating nonclassicality for weak bunched sources of light and show that our PDC source strongly violates this criterion, whereas it does not violate Hillery’s inequalities. Our violation of nonclassicality is the first time that nonclassical light has been unambiguously observed by direct photon number detection without any need for assumptions about the source.

Furthermore, the relatively innocuous assumption that detector inefficiency is due to linear loss processes enables us to reconstruct from raw data a photon number distribution that exhibits even-odd photon number oscillations, which strongly violate Hillery’s criteria. Although measurement of photon number oscillations have been reported [19], they have been inferred by performing optical homodyne tomography and thus involve assumptions to process the data; in contrast our reconstruction of photon number oscillations requires no assumptions about the source and only one assumption about the loss mechanism for detectors.

We first present a new test for nonclassical statistics. With an ideal (i.e., perfect efficiency) photon number detector, direct observation of PDC output is predicted to exhibit photon number oscillations [2]. For example, with \( P_n \) the probability of observing \( n \) photons, an ideal detector should yield \( P_1 = P_3 = 0 \) and \( P_2 > 0 \). However, an imperfect detector described by a linear loss model may not directly observe photon number oscillations, and thus we must establish a criterion for photon number statistics to be nonclassical. Define \( \Gamma \) as the ratio

\[
\Gamma = \frac{P_2}{P_1 + P_2 + P_3},
\]

(1)

For a perfect detector measuring PDC output, we predict \( \Gamma = 1 \).

We now prove that any semiclassical theory of light which is constrained to distributions of Poissonian photon
statistics cannot yield states with $\Gamma$ greater than a maximum classical bound $\Gamma_{\text{classical}}$. Semiclassical light has a Glauber-Sudarshan $P$ representation $P(\alpha)$ that obeys the requirements of a proper distribution [20], and the corresponding photon number distribution for semiclasical light can be written as a sum or integral of Poisson number distributions [21]. For a Poisson photon number distribution given by $P_n(\tilde{n}) = e^{-\tilde{n}} \tilde{n}^n/n!$, this ratio has a maximum value $\Gamma_{\text{classical}} = 3/(3 + 2\sqrt{6}) \approx 0.380$, saturated by the Poisson distribution with average photon number $\tilde{n} = \sqrt{6}$. However, one can show that this optimal value holds not only for a Poisson distribution, but for any classical distribution of Poissonians. First, we define the functions

$$f(\tilde{n}) = P_2(\tilde{n}), \quad g(\tilde{n}) = P_1(\tilde{n}) + P_2(\tilde{n}) + P_3(\tilde{n}).$$

Thus, for a Poisson distribution,

$$\Gamma(\tilde{n}) = \frac{f(\tilde{n})}{g(\tilde{n})} \leq \Gamma_{\text{classical}}.$$  

Now, consider a distribution of Poissonians with $P_n(\Phi) = \int d\tilde{n} \Phi(\tilde{n}) P_n(\tilde{n})$, where $\Phi(\tilde{n})$ is a probability distribution over $\tilde{n} \in \mathbb{R}^+$ satisfying $\int d\tilde{n} \Phi(\tilde{n}) = 1$. Calculating $\Gamma(\Phi)$ for this distribution of Poissonians gives

$$\Gamma(\Phi) = \frac{\int d\tilde{n} \Phi(\tilde{n}) f(\tilde{n})}{\int d\tilde{n} \Phi(\tilde{n}) g(\tilde{n})} = \frac{\int d\tilde{n} \Phi(\tilde{n}) g(\tilde{n})}{\int d\tilde{n} \Phi(\tilde{n}) g(\tilde{n})} \leq \frac{\int d\tilde{n} \Phi(\tilde{n}) g(\tilde{n})}{\int d\tilde{n} \Phi(\tilde{n}) g(\tilde{n})} = \Gamma_{\text{classical}},$$

where the inequality follows from Eq. (3) and the fact that $\Phi(\tilde{n})$ is non-negative and $g(\tilde{n})$ is strictly positive. Thus, no distribution of Poissonians can result in $\Gamma > \Gamma_{\text{classical}}$.

All classical light fields will lead to statistics that can be expressed as a distribution of Poissonians of photon number states. Thus, the classical theory of light predicts that the inequality

$$\Gamma \leq \Gamma_{\text{classical}} = 3/(3 + 2\sqrt{6})$$

cannot be violated. In contrast, one expects that light from PDC will lead to a violation of this condition, which can be demonstrated by simply measuring $P_1, P_2,$ and $P_3$.

In the presence of imperfect detection efficiency, loss may serve to degrade $\Gamma$ and a violation of the classical criterion may not be observed. Consider a PDC experiment in which the pump is sufficiently weak that the probability of generating more than one-photon pair is very small. In this case the ratio in Eq. (1) is given by $\Gamma = \eta/(2 - \eta)$, where $\eta$ is the detection efficiency. A violation of the inequality is not predicted unless $\eta \geq 3/7 \approx 0.55$.

Fortunately, the VLPC has the capability to detect photon number states with high quantum efficiency [12,13]. The VLPC has been shown to have quantum efficiencies approaching 90%. Furthermore, if more than one photon is incident on the detector surface, the height of the output electrical pulse is proportional to the number of incident photons. This gives us information about the number of photons that have been detected.

The experimental setup for testing nonclassical statistics is shown in Fig. 1. We use the fourth harmonic (266 nm) of a $Q$-switched neodymium-doped yttrium aluminum garnet (Nd:YAG) laser (20 ns pulse duration at 45 kHz repetition rate). Using a pulsed pump allows us to eliminate the detector dark counts (20000 s$^{-1}$) by temporal gating. The laser pumps a beta-barium-borate (BBO) crystal set for collinear degenerate type I phase matching (optic axis $47.6^\circ$ from the pump). In this configuration, the down-converted photons have half the energy of the pump (532 nm) and travel in the same direction. The pump is removed by a prism, while the down-conversion is focused by a 250 mm lens onto the VLPC detector. We also have the option of directly illuminating the detector with second harmonic light (532 nm) from the laser, which is a classical light source. The VLPC is held in a He bath cryostat at a temperature of 6.5 K. The detector output is amplified by low noise rf amplifiers. The signal to noise ratio of the output pulses is measured to be 27. The amplified electrical pulses are sent to a gated boxcar integrator, which is triggered by the laser. The boxcar integrates the pulse over a 20 ns window, and the output is sent to an analog-to-digital (A2D) converter and stored on a computer.

The output of the detector illuminated by light from the second harmonic of the laser is shown in Fig. 2. The pulse area spectrum features a series of peaks representing the different photon number state detections. In the inset we show the probability distribution, which is calculated by fitting each peak to a Gaussian function [22]. The area under each Gaussian curve gives the number of events representing that photon number. The area of each peak can be normalized by the total area to give the probability distribution, which appears as a Poisson distribution.

Figure 3(a) shows the pulse area histogram when the detector is illuminated by parametric down-conversion, using a pump power of 1 $\mu$W. At this weak pump intensity, a single pump pulse will usually generate zero
photos, while a photon pair is generated with a small probability. The probability of generating more than one-photon pair is very small. The figure focuses on the 1, 2, and 3 photon detection peaks, which we use to verify nonclassical statistics. The calculated probability distribution is shown in the inset. One can see that the probability of 1 and 2 photon detection is nearly equal, but the probability of 3 photon detection is nearly zero. These probabilities are \( P_1 = 0.0818 \), \( P_2 = 0.0696 \), and \( P_3 = 0.0061 \), which yields \( \Gamma = 0.442 \), representing a 40 standard deviation violation of the classical limit. In contrast, Hillery’s criteria [14] (namely, that \( P_{\text{even}} \equiv \sum_n P_{2n} < \frac{1}{2} \) or \( P_{\text{even}} - P_0 \geq \sum_n P_{2n+1} \equiv P_{\text{odd}} \)) are not violated, as our data yield \( P_{\text{even}} - P_0 = 0.061 \) and \( P_{\text{odd}} = 0.0702 \).

The large one-photon probability is due to losses from the detector and collection optics. In the limit of low excitation, the one- and two-photon probability can be used to calculate the detection efficiency, given by

\[
\eta = \frac{2 \bar{P}_1}{1 + 2 \bar{P}_1},
\]

From the measurements, it is calculated that the detection efficiency is 0.67. Using the measured VLPC quantum efficiency of 0.85, the photon collection efficiency is calculated to be 0.79.

Figure 3(b) shows the measured value of \( \Gamma \) as a function of pumping intensity. The black line represents the classical limit, which is violated for a large range of pumping intensities. At high pumping intensities \( \Gamma \) begins to drop. This drop is due to an increase in the two pair creation probability, which, in the presence of losses, will enhance the three photon detection probability. \( \Gamma \) also drops at low pumping intensities due to the dark counts of the VLPC which enhances the one-photon probability.

With high detector efficiency, the emitted output of PDC is predicted to feature even-odd photon number oscillations due to the two-photon nature of the process. These oscillations lead to the nonclassical statistics discussed in the previous section. Direct observation of these oscillations using the photon counting capability of the VLPC would be a remarkable achievement; unfortunately, such direct observation requires extremely high quantum efficiencies. Even the relatively high detection efficiencies of 0.67 in our experiment are not predicted to observe this oscillatory behavior. However, one can make an accurate independent measurement of the photon detection efficiency and correct for this effect in the photon number distribution. This allows the reconstruction of the original even-odd oscillations of the field.

The detection efficiency can be compensated as follows. Define \( p_i \) as the probability that the photon field contained \( i \) photons and \( f_i \) as the probability that \( i \) photons are detected. In the presence of losses and dark counts, these two distributions are related by

\[
f_i = \sum_{k=0}^{i} e^{-d} \frac{d^k}{k!} \sum_{j=i-k}^{\infty} \left( \frac{j}{i-k} \right) \eta^{i-k} (1 - \eta)^{j-(i-k)} p_j,
\]

where \( \eta \) is the detection efficiency and \( d \) is the average dark count rate in the integration window. In order to calculate \( p_i \) from \( f_i \), the above transformation must be inverted. To perform this inversion, we truncate the photon number distribution at some photon number \( n \), which is sufficiently large such that \( p_{n+1} = 0 \) is a good approximation. Under this approximation, the initial and
final probability distributions are simply related by a matrix, whose coefficients are given by Eq. (7). This matrix can be numerically inverted. It is important to emphasize that there are no fitting parameters in this model. The only two parameters, the quantum efficiency and dark counts of the VLPC, are both independently measured. Once they are known there is a one-to-one relationship between the actual and measured photon number distribution.

Figure 4 shows the result of the photon number reconstruction. Three different pumping intensities are used. The reconstructed photon number distribution, truncated at ten photons, is shown for each pumping intensity. The insets show the raw, uncorrected, number distributions. The reconstructed probabilities demonstrate very clear even-odd oscillations as predicted.

At higher photon numbers, it can be seen that the reconstructed distribution becomes slightly negative. This erroneous effect is caused by truncation error. As the pumping intensity is increased, the approximation that the photon distribution can be truncated after ten photons becomes less accurate. This error manifests itself in the probabilities becoming slightly negative for the nine and seven photon probability and is most pronounced at the largest pumping intensity of 8 μW, where the truncation approximation is least accurate. One could suppress this error by truncating at a higher photon number. Unfortunately, because of the limited range of our amplifiers and A2D converters, it is difficult to measure these higher order photon numbers in practice. This puts a limit on the pumping power one can use and still get a good reconstruction. It is possible that an improved numerical algorithm over simply putting a cutoff in the number distribution may overcome some of these practical difficulties.

In conclusion, we have directly observed nonclassical photon counting statistics from PDC. We have shown theoretically that the photon counting statistics for all classical fields must satisfy the inequality given in Eq. (5). Using the high quantum efficiency and photon number detection capability of the VLPC we have experimentally demonstrated violations of this inequality by light emitted from PDC. By correcting for the quantum efficiency and dark counts of the VLPC, we have also succeeded in reconstruction of the even-odd oscillation in the photon number distribution of light generated by the down-conversion field.

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FIG. 4 (color online). Reconstructed photon number distributions for 4, 6, and 8 μW pumping powers. The insets show uncorrected distributions.