Characterization of the nonclassical nature of conditionally prepared single photons

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(Received 22 December 2004; published 25 August 2005)

A reliable single photon source is a prerequisite for linear optical quantum computation and for secure quantum key distribution. A criterion yielding a conclusive test of the single photon character of a given source, attainable with realistic detectors, is therefore highly desirable. In the context of heralded single photon sources, such a criterion should be sensitive to the effects of higher photon number contributions, and to vacuum introduced through optical losses, which tend to degrade source performance. In this Rapid Communication we present, theoretically and experimentally, a criterion meeting the above requirements.

DOI: 10.1103/PhysRevA.72.021802

High fidelity single photon sources are an essential ingredient for quantum-enhanced technologies including linear optical quantum computation (LOQC) and secure quantum key distribution. Thus, the endeavor to generate single photons in controlled, well-defined spatio-temporal modes is an active area of research. Current single photon source candidates can be classified into two categories: deterministic sources producing single photons on demand at predefined trigger times and heralded single photon sources relying on the spontaneous emission of distinguishable photon pairs in conjunction with conditional preparation. While the emission times for conditional single photon sources cannot be controlled beyond the restriction of emission time slots through a pulsed pump, it has been shown that waveguided parametric down conversion (PDC) can yield heralded single photons in well-defined modes together with high collection efficiencies [1]. Conditional state preparation has been utilized in various physical systems including atomic cascades [2], ensembles of cold atoms [3], and in PDC. In the case of PDC, conditional preparation was first reported by Mandel et al. [4] and since then has been optimized to generate approximately n=1 Fock states [1,6–11]. In order to assess the performance of heralded single photon sources a criterion that takes into account the detrimental contributions of higher photon numbers and optical losses is needed. In addition, such a criterion should take into full consideration limitations of existing photodetectors such as the binary behavior of avalanche photodiodes operated in the Geiger mode where a single click signifies the detection of one or more photons. In this paper we derive such a criterion and show that our previously reported waveguided PDC source [1] represents a high fidelity source of heralded single photons.

A standard approach used to determine whether a light source exhibits classical or quantum photon statistics is the measurement of the $g^{(2)}(\tau)$ second-order intensity autocorrelation function in a Hanbury-Brown Twiss geometry. The semiclassical theory of photodetection predicts, firstly, that $g^{(2)}(0) \ge g^{(2)}(\tau)$ for all time delays τ , and, secondly, that $g^{(2)}(0) \ge 1$. The observation of photon antibunching, i.e., $g^{(2)}(0) \le g^{(2)}(\tau)$, has been utilized, for example, to verify the nonclassical character of deterministic single photon sources implemented by strongly coupled atom cavity systems [5]. For PDC sources, the probability of generating simultaneously two photon pairs at a given instant of time is of the

PACS number(s): 42.50.Ar, 03.67.Lx

same order as the probability of generating two independent pairs separated by the interval τ . This obliterates the effect of antibunching, unless we employ selective heralding that identifies specifically a single-pair component. For PDC sources, the nonclassical character of the generated radiation is usually tested by violating the lower bound on the secondorder intensity autocorrelation function $g^{(2)}(0) \ge 1$. $g^{(2)}(0)$ constitutes a figure of merit which determines the degree to which higher photon number contributions degrade the single photon character [9].

Based on a classical wave description and intensity measurements, Grangier *et al.* derived from the Cauchy Schwarz inequality a similar "anticorrelation" criterion for characterizing conditionally prepared single photons by coincidence detection rates [2]. For the experimental configuration in Fig. 1, an anticorrelation parameter

$$\alpha = R_1 R_{123} / R_{12} R_{13} \tag{1}$$

can be defined, which indicates nonclassical photon statistics for $\alpha < 1$, where R_i represents the singles count rates at detector *i*, and R_{ij} , R_{ijk} the double and triple coincidences for the respective detectors *i*, *j*, *k*. A variant of the $g^{(2)}(0)$ measurement specifically designed to study conditional single photon sources independently of losses, which was pioneered by Clauser [12], has recently been implemented for single photons generated from an ensemble of cold atoms [3].

In the works cited above, the theoretical modeling of experimental data was carried out in terms of intensity correlation functions. In a typical experiment, however, the count rates are directly related to light intensities only under certain



FIG. 1. Schematic of experimental setup designed to test the single photon character of a conditionally prepared state.

auxiliary assumptions. The reason for this is that standard photodetectors sensitive to single photons, such as avalanche photodiodes (APD) operated in the Geiger regime, do not resolve multiphoton absorption events and yield only a binary response telling us whether at least one photon was present in the detected mode or none at all. With such detectors, the light intensity can be read out from the count rates only in the limit of weak fields, where the probability of detecting a single photon is proportional to the intensity. In a general case, the probability of obtaining a click is a nonlinear function of the incident intensity. This aspect is particularly important in schemes utilizing ultrashort pulses, where the incoming light energy is concentrated in sub-picosecond time intervals that cannot be resolved even by the fastest photodetectors. It is therefore interesting to go beyond the basic intensity correlation theory and examine whether count statistics collected with binary non-photon-resolving detectors can serve as a test of source nonclassicality. We will demonstrate in the following that this is indeed the case. Furthermore, the nonclassicality criterion based on measuring $g^{(2)}(0)$ relies on a coincidence basis measurement so that losses can be neglected. However, for applications such as cascaded logic gates in LOQC [13] and loophole free tests of Bell inequalities [14], post-selection is not desirable, as it leads to vacuum contamination. The latter diminishes the usability of the single photon states: heralding no longer necessarily corresponds to the successful generation of a singlephoton or LOQC gate operation. In this paper we derive a criterion designed to test the nonclassical nature of conditionally prepared single photon states. Our criterion takes into account both the nonlinearity of the detectors, and the fidelity of the generated single-photon state, which measures the probability that a single photon is actually present when it is heralded. The criterion can be tested in a standard setup in which the signal field is subdivided into two submodes, each monitored by a nonphoton number resolving detector.

Consider a source emitting pairs of ultrashort light pulses, whose intensities integrated over the pulse durations and detector active area, are W_A and W_B . In the semiclassical theory of photodetection we will treat these quantities as nonnegative stochastic variables described by a joint probability distribution $\mathcal{P}(W_A; W_B)$. Beam B is divided by a beam splitter with power reflection and transmission coefficients r and t. Finally, the resulting beams are detected by three photodetectors. We will assume that the probability of obtaining a click on the *i*th detector illuminated by flux W is given by $p_i(W)$, bounded between 0 and 1. We furthermore assume $p_i(W)$ to increase monotonically with increasing light pulse energy W (an assumption which holds if saturation and nonlinear effects in the photodiode are avoided). Under these assumptions it is easy to show that the following inequality is satisfied for an arbitrary pair of arguments W_B and W'_B :

$$[p_2(rW_B) - p_2(rW'_B)][p_3(tW_B) - p_3(tW'_B)] \ge 0.$$
(2)

Indeed, the sign of both the factors in square brackets is always the same, depending on the sign of the difference $W_B - W'_B$; their product is therefore never negative. Let us now multiply both sides of the above inequality by the factor $\mathcal{P}(W_A; W_B)\mathcal{P}(W'_A; W'_B)p_1(W_A)p_1(W'_A)$ which is likewise non-negative, and perform a double integral $\int_0^\infty dW_A dW_B \int_0^\infty dW'_A dW'_B$. This yields the inequality

$$\mathcal{B}R_1^2 = R_1 R_{123} - R_{12} R_{13} \ge 0, \tag{3}$$

where the single, double, and triple count rates are given by averages $\langle ... \rangle = \int_0^\infty dW_A dW_B \mathcal{P}(W_A; W_B)...$ defined with respect to the probability distribution $\mathcal{P}(W_A; W_B)$:

$$\begin{split} R_1 &= \langle p_1(W_A) \rangle, \quad R_{12} = \langle p_1(W_A) p_2(rW_B) \rangle, \\ R_{13} &= \langle p_1(W_A) p_3(tW_B) \rangle, \quad R_{123} = \langle p_1(W_A) p_2(rW_B) p_3(tW_B) \rangle. \end{split}$$

It is seen that the inequality derived in Eq. (3) which can be transformed into

$$R_1 R_{123} / R_{12} R_{13} \ge 1 \tag{4}$$

has formally the same structure as the condition derived by Grangier *et al.* [2]. However, the meaning of the count rates is different, as we have incorporated the binary response of realistic detectors. It is noteworthy that this inequality has been derived with a very general model of a detector, assuming essentially only a monotonic response with increasing light intensity.

Our experimental apparatus is similar to that reported in Ref. [1]. PDC is generated by a KTiOPO₄ (KTP) nonlinear waveguide pumped by femtosecond pulses from a modelocked, frequency doubled 87 MHz repetition rate Ti:sapphire laser. The average repetition rate in the generated PDC $(\sim 70 \text{ kHz})$ is small enough that saturation effects due to detector dead time are largely avoided. In contrast to that reported in Ref. [1], the approach here is to record timeresolved detection information for the three spatial modes involved with respect to the Ti:sapphire pulse train as detected by a fast photodiode. We thus obtain a reference clock signal with respect to which post-detection event selection can be performed in order to implement temporal gating. The latter is important for the suppression of uncorrelated background photons, the presence of which can lead to heralded vacuum (rather than a true single photon). Through this approach, we are able to freely specify the time-gating characteristics; arbitrarily complicated logic can be performed without added experimental hardware. Drawbacks include the lack of real-time data processing as well as the dead time in the region of μ s between subsequent triggers exhibited by the digital oscilloscope (LeCroy WavePro 7100) used for data acquisition. In our setup, source brightness information is obtained via a separate NIM electronics-based measurement.

For a given trigger event, three numbers are recorded: the time difference between the electronic pulse positive edge corresponding to the trigger and to the two signal modes t_{S1} , t_{S2} , as well as the trigger-clock reference time difference t_{CLK} . Time gating involves discarding trigger events outside a certain range of t_{CLK} values, while coincidence events with t_{S1} and t_{S2} outside a 1.1 ns wide coincidence window are regarded as accidental and ignored. We collected 75000 trigger events and measured detection efficiencies before time gating (defined as the rate of coincidences normalized by singles) for each of the two signal channels of 14.4% and 13.7%. Figure 2 shows the post-processed data using a scanned temporal band-pass filter with 300 ps width (se-



PHYSICAL REVIEW A 72, 021802(R) (2005)

FIG. 2. (Color online) (a) Time-resolved doubles and singles count rates for signal₁. (b) Time-resolved doubles and singles count rates for signal₂. (c) Conditional detection efficiency for signal₁. (d) Conditional detection efficiency for signal₂. (e) Time-resolved triple count rate. (f) Time-resolved inequality parameter exhibiting violation.

lected to approximately match the measured APD jitter; for graphical clarity, the time difference between displayed data points is less than the filter width used). Figures 2(a) and 2(b) shows the time-resolved signal₁-trigger [signal₂-trigger] coincidence count rate, compared to the time-resolved trigger singles count rate. Figures 2(c) and 2(d) shows the detection efficiency resulting time-gated for the signal₁ [signal₂] channel, showing maximum values of $\sim 17.4\%$ [$\sim 17.0\%$]. Figure 2(e) shows time-resolved triple coincidences, for identical coincidence windows as used in computing double coincidences. Thus, our time-gating procedure filters the PDC flux so that for the pump power used, the generated light is described essentially by a superposition of vacuum with single photon pairs, showing nearly vanishing multiple pair generation.

Figure 2(f) shows the time-resolved inequality parameter [see Eq. (3)] resulting from the count rates presented above. For example, at the triples counts peak, we obtain the following time-gated counting rates for 75000 trigger events: $R_{123}=2$, $R_{12}=5329$, $R_{13}=5067$, and $R_{1}=30629$, yielding an inequality parameter value of $\mathcal{B}=-0.029\pm.001$. For comparison, our results correspond to a value of the anticorrelation parameter of $\alpha = (2.3 \pm 1.6) \times 10^{-3}$. Figure 2 indicates an overall signal transmission [defined as the sum of the two individual efficiencies $(R_{12}+R_{13})/R_1$] of ~34.5%. The main

contribution to losses is the nonunit quantum efficiency of the single photon detectors. The overall detection efficiency is also degraded due to imperfect optical transmission and remaining unsuppressed uncorrelated photons. From the above count rates, we can also calculate $g^{(2)}(0) = 2p_{(2)}/p_{(1)}^2$ in terms of the probability of observing a single photon in the signal arm $p_{(1)} = (R_{12} + R_{13})/R_1$ and the probability of observing two photons in the signal arm $p_{(2)} = R_{123}/R_1$. We thus obtain $g^{(2)}(0) = (1.1 \pm 0.8) \times 10^{-3}$, amongst the lowest reported for a single photon source.

Ignoring the spectral and transverse momentum degrees of freedom, the signal and idler photon-number distribution in a realistic PDC source is expressed as

$$|\Psi\rangle = \sqrt{1 - |\lambda|^2} \sum_{n=0}^{\infty} \lambda^n |n\rangle_s |n\rangle_i, \tag{5}$$

where *n* represents the photon number describing each of the signal and idler modes and λ represents the parametric gain. PDC experiments often operate in a regime where λ is small enough that the probability of multiple pair generation becomes negligible. For larger values of λ (accessed, for example, by a higher pump power or higher nonlinearity), however, the higher order terms (e.g., $|2\rangle_s |2\rangle_i, |3\rangle_s |3\rangle_i...$) become important. While these higher photon number terms are de-



FIG. 3. (a) Inequality parameter \mathcal{B} as a function of the overall transmission in the signal arm (for fixed λ values as shown). (b) Inequality parameter \mathcal{B} as a function of the parametric gain (for fixed η values as shown). Black squares represent the experimentally obtained value for the inequality parameter assuming λ =0.03; the uncertainty is smaller than the square dimensions. A trigger detection efficiency η_T =0.02 was assumed.

sirable for conditional preparation via photon number resolving detection, in the context of the present work, where the detectors used are not photon-number resolving and where the goal is high-fidelity preparation of *single* photons, multiple pair generation must be avoided. As discussed earlier, in order to characterize a source of conditionally prepared single photons based on PDC, besides the parametric gain λ , optical losses must be taken into account. Losses in the signal arm imply that a trigger detection event can incorrectly indicate the existence of a signal photon, while in reality vacuum is present. Figure 3 shows the expected inequality behavior based on a quantum mechanical calculation where we assume that the detection probability is given by the expectation value of the operator $1 - \exp(-\eta \hat{W})$ (where \hat{W} is the time-integrated intensity operator and η is the corresponding overall transmission including all optical and detection losses) [15,16]. Figure 3(a) shows the calculated inequality parameter \mathcal{B} [see Eq. (3)] for PDC light as a function of the overall signal optical transmission $\eta_s = (R_{12} + R_{13})/R_1$ for a fixed value of the parametric gain λ . Figure 3(b) shows the inequality coefficient as a function of the parametric gain λ for different levels of optical loss. Note that a strong violation of the inequality is only observed in the low parametric gain limit coupled with low losses. Note further that the minimum value of \mathcal{B} , corresponding to the strongest violation and which is only reached in the ideal lossless case, is -0.25. In an experimental realization, while accessing very low values of λ is straightforward, e.g., by using a low pump

PHYSICAL REVIEW A 72, 021802(R) (2005)

power, attaining a sufficiently low level of loss to yield a nearly ideal violation is challenging. Under the assumption that all uncorrelated trigger photons are suppressed, it may be shown that λ is given in terms of experimentally measurable quantities as

$$\lambda^2 = (R_2 + R_3) / \eta_s R_{rep} (1+f), \tag{6}$$

where R_{rep} is the pump repetition rate and f is the uncorrelated photon intensity normalized by that of PDC. We estimate that in our experiment f fulfills $0 < f \le 2$. Our experimental values of $R_2 + R_3 \approx 70000 \text{ s}^{-1}$, $R_{rep} = 87 \times 10^6 \text{ s}^{-1}$ and $\eta_s = 0.345$ thus yield $0.016 < \lambda < 0.047$. The experimentally observed violation [see Fig. 2(f)] is in good agreement with the theory curves in Fig. 3. The black squares in Figs. 3(a) and 3(b) depict the observed violation as compared with the theoretical curves, where the uncertainty is smaller than the square dimensions. The plot in Fig. 3(a) assumes a fixed value of the parametric gain λ (different curves shown for a choice of λ values). The signal arm transmission η_s is obtained as the sum of the two individual signal detection efficiencies [see Figs. 2(a) and 2(b)].

In summary, we have derived a criterion which allows a conclusive test of the single photon character of conditionally prepared single photon states. We have shown that the inequality in Eq. (3) is fulfilled by all classical light sources, as well as by states generated by PDC exhibiting higher photon numbers through a large parametric gain. On the contrary, a strong violation of the inequality is observed only for states that constitute a good approximation to a conditionally prepared single photon. Our criterion is realistic enough to include binary nonphoton number resolving photon counting detectors while it is sensitive to the degradation observed in the prepared state caused by a vacuum component due to losses. Application of our criterion shows that our waveguided PDC source [1] constitutes a high-fidelity conditional single photon source. Our derived inequality yields a new figure of merit quantifying the overall performance of conditional single photon sources taking into consideration key experimental imperfections.

This work was supported by ARDA Grant No. P-43513-PH-QCO-02107-1. A.U. acknowledges support from the Center for Quantum Information, funded by ARO administered MURI Grant No. DAAG-19-99-1-0125.

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