## <span id="page-0-1"></span>Correlated Multiphoton Holes: Absence of Multiphoton Coincidence Events

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We generate bipartite states of light which exhibit an *absence* of multiphoton coincidence events between two modes amid a constant background flux. These ''correlated photon holes'' are produced by mixing a coherent state and relatively weak spontaneous parametric down-conversion by using a balanced beam splitter. Correlated holes with arbitrarily high photon numbers may be obtained by adjusting the relative phase and amplitude of the inputs. We measure states of up to five photons and verify their nonclassicality. The scheme provides a route for observation of high-photon-number nonclassical correlations without requiring intense quantum resources.

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Introduction.—The generation of multiphoton entangled states has motivated a large body of experimental work in quantum optics [[1](#page-3-1)[–3](#page-3-2)]. The workhorse in such experiments has been spontaneous parametric down-conversion (SPDC), which allows generation of entangled photon pairs [[4](#page-3-3)]. In recent years there has been an ongoing effort to create nonclassical states with more and more photons, the highest value being a six-photon graph state [\[5\]](#page-3-4) using three SPDC pairs. Scaling up the number of photons in such schemes, however, is challenging since they rely on multiple emissions of SPDC pairs. This could be accomplished by using state-of-the-art, high intensity sources to pump the SPDC [\[6](#page-3-5)].

Recently, the inverse of SPDC, namely, the process in which a photon pair is *missing* amid a constant background flux, has been demonstrated experimentally [[7](#page-3-6)]. This state has been dubbed an ''entangled photon hole'' and, just like regular entangled photons, it can be used to violate Bell's inequality [[8](#page-3-7)]. Here we generalize this concept to more than two photons by creating two-mode states in which the probability for arbitrary photon numbers  $N_1$  and  $N_2$  to arrive simultaneously in the respective modes is zero, where choosing  $N_1 = N_2 = 1$  corresponds to the case of entangled photon holes [[7](#page-3-6)[,8\]](#page-3-7). We refer to the generated states as ''correlated photon holes'' (CPHs). As in the twophoton case [\[7](#page-3-6)], our scheme involves the mixing of SPDC and coherent light. Interestingly, the larger  $N_1$  and  $N_2$ , the higher the relative weight of the coherent light, implying that our scheme may be implemented at high photon numbers with very modest SPDC fluxes.

Theoretical scheme.—To date, a handful of photoncounting experiments have utilized interference of coherent light and SPDC in a configuration sensitive to the relative phase. The SPDC in these was produced in either a single pass geometry [[7](#page-3-6)[,9–](#page-3-8)[11](#page-3-9)] or an optical parametric oscillator [\[12\]](#page-3-10). With the exception of our recent demonstration of ''high-NOON'' states [[9](#page-3-8)], these experiments focused on two-photon correlations. Here we generate another class of high-photon-number states which emerges naturally in this type of interference.

Consider a  $50/50$  beam splitter fed by a coherent state  $|\alpha\rangle_a$  in one input port and collinear degenerate SPDC  $|\xi\rangle_b$  $\alpha/a$  in one input port and confined degenerate SFDC  $\frac{1}{5}/b$ <br>in the other [see Fig. [1\(a\)\]](#page-1-0). The input states are defined in the conventional way [[13](#page-3-11)]:

$$
|\alpha\rangle = \sum_{n=0}^{\infty} e^{-(1/2)|\alpha|^2} \frac{\alpha^n}{\sqrt{n!}} |n\rangle, \qquad \alpha = |\alpha| e^{i\phi_{cs}},
$$
  

$$
|\xi\rangle = \frac{1}{\sqrt{\cosh r}} \sum_{m=0}^{\infty} (-1)^m \frac{\sqrt{(2m)!}}{2^m m!} (\tanh r)^m |2m\rangle,
$$
 (1)

where the phase of  $\ket{\xi}$  has been set arbitrarily to zero,<br>leaving the relative phase of the two inputs to be deterleaving the relative phase of the two inputs to be determined by  $\phi_{\text{cs}}$ . We denote the pair amplitude ratio of the coherent state and SPDC by

$$
\gamma \equiv |\alpha|^2 / r. \tag{2}
$$

<span id="page-0-0"></span>In physical terms  $\gamma^2$  is the two-photon probability of the classical source divided by that of the quantum source (in the limit r,  $|\alpha| \ll 1$ ). The larger the value of  $\gamma$ , the higher the relative weight of the classical resources. We denote the path entangled state at beam splitter (BS) output modes c and d [Fig. [1\(a\)](#page-1-0)] by  $|\psi_{\text{out}}\rangle_{c,d}$ . The amplitude for  $N_1$  and  $N_2$ photons simultaneously in the BS output modes is then given by

$$
A_{N_1,N_2}(\alpha, r) = \langle N_1, N_2 | \psi_{\text{out}} \rangle_{c,d}.
$$
 (3)

In the absence of SPDC this is simply

$$
A_{N_1,N_2}(\alpha,0) = e^{-|\alpha/\sqrt{2}|^2} (\alpha/\sqrt{2})^{N_1+N_2}/\sqrt{N_1!N_2!},\qquad(4)
$$

which is nonzero for all values of  $N_1$  and  $N_2$ . By adding SPDC  $(r > 0)$  it is possible to cancel  $A_{N_1,N_2}$  for arbitrary values of  $N_1$  and  $N_2$  by correctly adjusting  $\gamma$  and  $\phi_{\text{cs}}$ . In the limit  $r \ll 1$ , there are  $\lfloor (N_1 + N_2)/2 \rfloor$  distinct solutions for the equation  $A_{N_1,N_2} = 0$  for any choice of  $N_1$  and  $N_2$ . For example, for  $N_1$ ,  $N_2 = 1$ , 1 we choose  $\gamma = 1$  and  $\phi_{cs} =$  $\pi/2$ . This produces an absence of 1, 1 coincidence events at the BS outputs which is similar to the previously studied case of two-photon holes [[7\]](#page-3-6). As a rule, the higher the values of  $N_1$  and  $N_2$ , the higher the value of  $\gamma$ , implying

<span id="page-1-1"></span>

<span id="page-1-0"></span>FIG. 1 (color online). Experimental setup for generation of correlated photon holes. (a) Schematic of the setup depicting a  $50/50$  beam splitter fed by a coherent state and SPDC. The correlated photon holes occur in modes  $\hat{c}$  and  $\hat{d}$  after the beam splitter by adjusting the relative phase and amplitude of the two input beams. Measurement of multiphoton coincidences is performed by using photon-number-resolving detectors. (b) Detailed layout of the setup. A pulsed Ti:sapphire oscillator with 120 fs FWHM pulse width and 80 MHz repetition rate is doubled by using a 2.74 mm lithium triborate (LBO) crystal to obtain 404 nm ultraviolet pulses with maximum power of 225 mW. These pulses then pump collinear degenerate type-I SPDC at a wavelength of 808 nm by using a 1.78 mm long beta barium borate (BBO) crystal. The SPDC ( $\hat{H}$  polarization) is spatially and temporally overlapped with a coherent state  $(\hat{V})$ polarization) by using a PBS. A thermally induced drift in the relative phase,  $\phi_{\text{cs}}$ , between  $\hat{H}$  and  $\hat{V}$  polarizations is corrected every few minutes by using a LC phase retarder. The coherent light amplitude is adjusted by using a variable attenuator. Modes  $\hat{a}$  (b) of panel (a) are realized collinearly by using  $\hat{V}$  (H) polarizations, respectively. These modes are then coherently mixed by using a polarization maintaining fiber (PMF) aligned with the  $\pm 45^\circ$  ( $\hat{X}$ ,  $\hat{Y}$ ) polarization axes. The spatial and spectral modes are matched by using a (single-mode) PMF and a 3 nm FWHM bandpass filter (BPF) centered at 808 nm. Photonnumber-resolving detection is performed using five single-photon-counting modules (SPCM, Perkin Elmer). Additional components: long-pass filter (LPF) and short-pass filter (SPF).

that relatively more photons originate from the classical source than from the SPDC. In the experimental part we demonstrate the scheme by measuring two cases, a four-photon CPH with  $N_1$ ,  $N_2 = 2$ , 2 and  $\gamma^2 = 3$ and a five-photon CPH with  $N_1$ ,  $N_2 = 5$ , 0 and  $\gamma^2 =$  $[15/(5-\sqrt{10})]^2 \sim 66.58$ . In both cases, the overall photon flux contributed by the SPDC is negligible compared to the coherent state flux. The SPDC may therefore be viewed as a small perturbation to the coherent field. It is noteworthy that our scheme bears some resemblance to a theoretical proposition for observation of antibunching using a degenerate parametric amplifier [[14](#page-3-12)].

Experimental setup and results.—Our setup (Fig. [1\)](#page-1-1) is similar to the one we have used for generation of NOON states with high photon numbers in a recent experiment [[9\]](#page-3-8). The scheme requires generation of SPDC and coherent light with common spatial and spectral modes. The beams are prepared in perpendicular linear polarizations  $(\hat{H}, \hat{V})$ and overlapped by using a polarizing beam-splitter cube (PBS). The phase between the beams,  $\phi_{\text{cs}}$ , is controlled before the PBS by using a liquid crystal (LC) phase retarder. The BS of Fig. [1\(a\)](#page-1-0) is then implemented in a collinear geometry by using a polarization-maintaining fiber with axes aligned at  $\pm 45^\circ$  ( $\hat{X}$ ,  $\hat{Y}$ ). Multiphoton coincidences with  $N_1$  and  $N_2$  photons arriving simultaneously in detectors  $D_1$  and  $D_2$ , respectively, are measured as a function of  $\phi_{\rm cs}$  by using an array of avalanche photodiodes. First, we create a four-photon hole by choosing  $N_1, N_2 = 2, 2$  (Fig. [2\)](#page-1-2); i.e., the simultaneous arrival of two photons in  $D_1$  and two photons in  $D_2$  is canceled while keeping a constant background flux. For this case  $\gamma^2 = 3$ [see Eq. ([2](#page-0-0))]. Next, we create a five-photon hole by choos-

<span id="page-1-2"></span>

FIG. 2 (color online). Measurement of a 2, 2 correlated photon hole. (a) (left y axis) Coincidence events with two photons in  $D_1$ and two photons in  $D_2$  as a function of relative phase  $\phi_{cs}$ between the coherent state and the SPDC (see Fig. [1](#page-1-1)). The minima correspond to phases in which the 2; 2 coincidence is canceled, implying a correlated photon hole. The solid line is a theoretical calculation taking into account the overall setup transmission and detector positive-operator-valued measures [\[9\]](#page-3-8). The visibility is  $94.9 \pm 1\%$ . Error bars indicate  $\pm \sigma$  statistical uncertainty. The dashed line indicates the classical bound for this measurement; see Eq. ([6\)](#page-2-0) and the preceding discussion. All points below this line (shaded area) exhibit nonclassical behavior. The two arrows indicate the position of the correlated photon holes. (b), $(c)$  (right y axis) The single counts in detectors  $D_1$  and  $D_2$  as a function of  $\phi_{cs}$  which exhibit virtually no phase dependence; the straight solid line is a guide to the eye.

<span id="page-2-1"></span>

FIG. 3 (color online). Measurement of a 5, 0 correlated photon hole. Here, all five detectors were employed in  $D_1$  by placing an  $\hat{H}$  transmitting polarizer followed by a half-wave plate just before the PBS in the detector box. This allows us to measure five-photon events in  $D_1$  $D_1$  (see Fig. 1). (a) (left y axis) Coincidence events with five photons in  $D_1$  as a function of relative phase  $\phi_{\rm cs}$  between the coherent state and the SPDC. The minimum, indicated by an arrow, corresponds to the phase in which the  $N_1 = 5$ ,  $N_2 = 0$  coincidence is canceled, implying a correlated five-photon hole. The solid line is a theoretical calculation taking into account the overall setup transmission and detector positive-operator-valued measures [[9\]](#page-3-8). The visibility is 98.6  $\pm$  1.3%. Error bars indicate  $\pm \sigma$  statistical uncertainty. The dashed line indicates the classical bound for this measurement; see Eq. ([6](#page-2-0)) and the preceding discussion. All points below this line (shaded area) exhibit nonclassical behavior. (b) (right y axis) The single counts in detector  $D_1$  as a function of  $\phi_{cs}$  which exhibit virtually no phase dependence; the straight solid line is a guide to the eye.

ing  $N_1, N_2 = 5, 0$  (Fig. [3\)](#page-2-1); i.e., we cancel the probability for simultaneous arrival of five photons in  $D_1$  and zero in  $D_2$ . In practice, for this case, we used all five of our singlephoton detectors [Perkin Elmer single-photon-counting modules (SPCMs)] in  $D_1$  and therefore could not postselect events with zero counts in  $D_2$ . At the employed flux, however, the higher order 5, 1 coincidence events (which could have been eliminated by postselection) are extremely rare with only a minor effect on the measured signal and may be neglected. For the five-photon case,  $\gamma^2 = 66.58$ , implying that the coherent state two-photon probability is 66.58 times larger than that of the SPDC. Remarkably, such a weak perturbation of quantum light has a dramatic effect on the measured five-photon events. The single counts, on the other hand, show no phase dependence in either case as seen in Figs. [2](#page-1-2) and [3](#page-2-1) (triangles). This is because the SPDC process generates only even photon numbers with no single-photon events to interfere with those of the coherent state.

Because of losses, higher order events have the effect of adding a background to the N photon coincidence measurement. This reduces the experimental visibilities and is the main limiting factor in observing photon holes with higher photon numbers. The overall transmission in our setup, which accounts for detector efficiencies and other sources of loss, is 12.5%. This is measured by taking the ratio of coincidence to single counts using only the SPDC beam (coherent state blocked). The solid lines accompanying the experimental plots (Figs. [2](#page-1-2) and [3](#page-2-1)) are calculated by using an analytical model of the experiment taking into account the overall transmission and the detector positiveoperator-valued measures [[9\]](#page-3-8).

Nonclassical properties.—To quantify the nonclassicality of the generated states, we derive a classical bound for Glauber's nth order, equal-time, correlation function  $g^{(n)}(\tau = 0)$ . For  $\alpha \ll 1$  this correlation function is propor-<br>tional to the *n* photon coincidence signal of a single spatial tional to the  $n$  photon coincidence signal of a single spatial mode divided by the single count raised to the nth power. Note that we chose  $\tau = 0$  since we are interested in photons arriving to the detectors at the same time. For thermal light, which exhibits bunching,  $g^{(n)}(0) = n!$ , and for a coherent state  $g^{(n)}(0) = 1$  [13.15]. In the following we coherent state  $g^{(n)}(0) = 1$  [[13](#page-3-11),[15](#page-3-13)]. In the following, we show that the result for a coherent state is actually the show that the result for a coherent state is actually the lower bound for an arbitrary classical state, i.e., a state with an arbitrary non-negative well-behaved P function [[16\]](#page-3-14). The inequality was implied in Titulaer and Glauber's work on high-order correlation functions of coherent fields [\[16\]](#page-3-14) and is derived here somewhat differently:

<span id="page-2-2"></span>
$$
g^{(n)}(\tau=0) = \frac{\mathrm{Tr}\{\hat{\rho}\hat{E}^{(-)^n}\hat{E}^{(+)^n}\}}{\mathrm{Tr}\{\hat{\rho}\hat{E}^{(-)}\hat{E}^{(+)^n}\}} = \frac{\int P(\alpha)|\alpha|^{2n}d^2\alpha}{\left[\int P(\alpha)|\alpha|^2d^2\alpha\right]^n} \ge 1,
$$
\n(5)

where the last inequality follows immediately from a multidimensional form of Jensen's inequality [[17](#page-3-15),[18](#page-3-16)]. We note that for  $n = 2$  this inequality is a well known result of Cauchy's inequality [[13](#page-3-11)]. Correlated photon holes require a bound for the two-point equal-time Glauber correlation function  $g^{(m,n)}(x_1, x_2; \tau = 0)$ , where  $x_1$  and  $x_2$  correspond<br>to the BS output modes [Fig. 1(a)]. The derivation is based to the BS output modes [Fig. [1\(a\)\]](#page-1-0). The derivation is based on the single-mode result, Eq. ([5\)](#page-2-2), and requires the assumption that the two-mode  $P$  function describing the BS outputs is separable. i.e.,  $P_{1,2}(\alpha, \beta) = P_1(\alpha)P_2(\beta)$ , a condition which is satisfied by all classical two-mode Gaussian states [[19](#page-3-17)]. By using this assumption, the inequality follows immediately from the single-mode result applied to each of the modes independently:

<span id="page-2-0"></span>
$$
g^{(m,n)}(x_1, x_2; \tau = 0) = \frac{\operatorname{Tr}\{\hat{\rho}\hat{E}^{(-)^m}(x_1)\hat{E}^{(-)^n}(x_2)\hat{E}^{(+)^m}(x_1)\}}{\operatorname{Tr}\{\hat{\rho}\hat{E}^{(-)}(x_1)\hat{E}^{(+)^n}(x_1)\}^m \operatorname{Tr}\{\hat{\rho}\hat{E}^{(-)}(x_2)\hat{E}^{(+)}(x_2)\}}}
$$
  
= 
$$
\frac{\int P_{1,2}(\alpha, \beta)|\alpha|^{2m}|\beta|^{2n}d^2\alpha d^2\beta}{\left[\int P_{1}(\alpha)|\alpha|^{2}d^2\alpha\right]^{m}\left[\int P_{2}(\beta)|\beta|^{2}d^2\beta\right]^{n}} = \frac{\int P_{1}(\alpha)|\alpha|^{2m}d^2\alpha}{\left[\int P_{1}(\alpha)|\alpha|^{2}d^2\alpha\right]^{m}}\frac{\int P_{2}(\beta)|\beta|^{2n}d^2\beta}{\left[\int P_{2}(\beta)|\beta|^{2}d^2\beta\right]^{n}} \ge 1.
$$
 (6)

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The classical bounds in Figs. [2](#page-1-2) and [3](#page-2-1) are obtained by raising the single counts to the appropriate powers and multiplying by a numerical factor to account for the repetition rate and the structure of our detector [Fig. [1\(b\)\]](#page-1-0). For the four-photon case (Fig. [2](#page-1-2)) we substitute  $m = 2$  and  $n =$ 2, violating the classical bound by 23.92 and 21.17 standard deviations at the phases for which the photon holes are created. For the five-photon case (Fig. [3\)](#page-2-1) we substitute  $m = 5$  and  $n = 0$ , and the bound is violated by 25.5 standard deviations.

Conclusion.—Mixing coherent light and SPDC is typically done in conjunction with homodyne detection [\[13\]](#page-3-11) by using a ''macroscopic'' local oscillator as the coherent state. Adopting this paradigm in the ''few photon'' regime and using number-resolving detectors brings to light a rich structure exhibiting various nonclassical signatures. This enabled us to create NOON states [[9](#page-3-8)] in a recent work and correlated photon holes here. Extending the present work to higher photon numbers can be done by using essentially the same setup, requiring only additional detectors to enable higher coincidence measurements. This does not entail a larger SPDC flux since relatively more of the photons originate from the coherent state which is practically unlimited in intensity, providing experimental simplification. As in the case of NOON states, however, the visibility of interference is limited by the overall setup transmission (currently 12.5%), determined by accounting for all sources of photon loss including the detector efficiency. This value could be improved by using high purity SPDC sources which can be spectrally mode matched to coherent states without requiring a bandpass filter [[20](#page-3-18)–[22\]](#page-3-19). Improved single-mode fiber coupling of the photon pairs and high efficiency photon-number-resolving detectors [\[23](#page-3-20)[,24\]](#page-3-21) would also be beneficial. Reaching a transmission of 25% would allow measuring states with ten photons in a setup similar to ours.

In this work, we have focused on the elimination of specific multiphoton events due to interference of coherent and down-converted light. Such photon holes are intriguing since their nonclassicality stems from the absence of multiphoton events. Indeed, such states have been shown to be potentially useful in increasing the secure data rate and transmission distance in quantum cryptography [[25](#page-3-22),[26](#page-3-23)]. They may also be beneficial in the context of high precision quantum measurements [\[27\]](#page-3-24). It would be interesting to compare their performance with that of previously studied states, particularly in the presence of loss [[28](#page-3-25)].

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