Photon Antibunching in Pulsed Squeezed Light Generated via Parametric Amplification

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We have measured the intensity correlation of pulsed squeezed light generated by a degenerate optical parametric amplifier seeding weak coherent input signal. When the signal was deamplified, the correlation showed antibunching and the probability distribution of the photon number was sub-Poissonian, both of these being nonclassical properties of light.

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Recently there has been much interest in nonclassical properties of light, which are understandable only in terms of quantized electromagnetic field. These properties are readily revealed in the intensity correlation which is proportional to the probability of two photons being detected at a time interval of τ. Classical field can give an interpretation to the following two cases: the probability is maximum at $\tau = 0$ and decreases as τ increases (e.g., thermal light), and the probability has a constant value for all τ (coherent light). The former case is called "bunching." However, we need quantized electromagnetic field for cases where the probability at $\tau = 0$ is smaller than that for larger τ. This phenomenon is termed "antibunching." Experimentally, it has been observed in the fluorescence from a system of a very small number of atoms or molecules excited by cw coherent light [1–4]. It was also observed by the use of a detection-triggered optical shutter in parametrically down-converted light [5]. Another way to observe antibunching is to inject coherent light into an interaction region in which two photons are simultaneously absorbed. Multiphoton absorption [6,7], parametric amplification [8], and others [9] have been proposed for the interaction process, but observations of antibunching by these methods have not been previously carried out, to the best of our knowledge.

In this Letter, we report the first experimental observation of photon antibunching in pulsed squeezed coherent light generated via degenerate parametric amplification. The scheme we used is similar to that proposed by Stoler [8], but we employed pulsed light instead of cw light in order to obtain large nonlinear response even from a single-pass geometry. In addition, the use of the pulsed light has an advantage in that the effect of antibunching can be detected even if its correlation time is shorter than the detector response time [10].

Let us consider that the fundamental light and the second harmonic light of a cw mode-locked laser are injected to an optical parametric amplifier. The output field is beam split and shone on two detectors, and time intervals between two photon-counting events are measured. When the pulse duration of the incident light and the response time of the detector used are much shorter than the time interval between two successive pulses, the recorded data give normalized intensity correlation of pulses of the nth neighbor:

$$g_n^{(2)} \equiv \frac{\left\langle \mathcal{T} : \int \hat{I}(t)dt \int \hat{I}(t+nT)dt : \right\rangle}{\left\langle \int \hat{I}(t)dt \right\rangle \left\langle \int \hat{I}(t+nT)dt \right\rangle}, \tag{1}$$

where $\hat{I}(t)$ is the light intensity operator, the operator product is written in normal order and in time order, T is the time interval of the pulses, and the integrations run over the duration of a single pulse. Since bunching or antibunching means the tendency that more or fewer photon pairs are detected close together in time than further apart [11], we may say that bunching occurs when $g_0^{(2)} > g_n^{(2)}$ and antibunching occurs when $g_0^{(2)} < g_n^{(2)}$ ($n \neq 0$) in the situation considered here. The latter condition, like in the case for cw light, violates the Schwarz inequality for classical field, and therefore the antibunching requires a quantized-field picture for its interpretation.

Our experimental apparatus is shown in Fig. 1. A cw mode-locked Nd-doped yttrium aluminum garnet (Nd:YAG) laser (Spectron model ML903) generates lin-

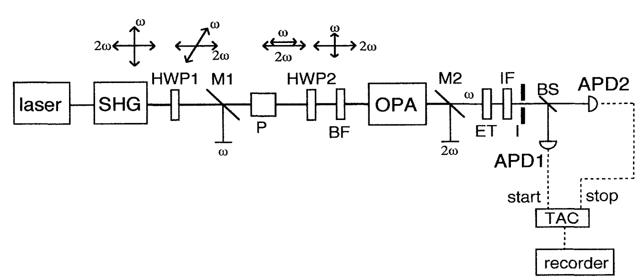

FIG. 1. The schematic drawing of the experimental apparatus; HWP1-2, harmonic wave plates; M1-2, mirrors; BF, birefringent filter; P, polarizer; OPA, parametric amplifier; BS, beam splitter; ET, etalon; IF, interference filter; I, iris; APD1-2, photon-counting avalanche photo diodes; TAC, time-to-amplitude converter. Arrows show the direction of polarization of the signal (ω) and the pump (2ω).

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early polarized infrared (IR) pulses at 1.064 μm with FWHM ~100 psec at a repetition rate of 82 MHz. The fundamental light is injected into a LiB_3O_5 crystal and the second harmonic light (~250 mW) is produced. The second harmonic is used as a pump field and the residual IR field as a signal for a single-pass degenerate optical parametric amplifier (OPA) made of a 5.8 mm long $KNbO_3$ crystal. The observability of antibunching in this experiment is crucially dependent on the stability of the relative phase between the signal and the pump. In order to stabilize the phase, the two beams are aligned collinearly. Harmonic wave plates and mirrors are used in order to adjust the intensity of the IR light independently. The harmonic mirrors (M1) reflect the IR light and make the signal field extremely weak ($\sim 10^{-11}$ W). The harmonic wave plate (HWP1) serves as a $\lambda/2$ plate for the 1.064 μm IR light and as a λ plate for the 532 nm green light, and rotates the direction of polarization of the IR light alone. Thus the combination of HWP1 and a polarizer (P) allows us to change the intensity of the signal field while leaving that of the pump unchanged. After that, the polarizations of the two fields are made orthogonal to each other again by another harmonic wave plate (HWP2), and their relative phase is adjusted with a birefringent filter (BF) by tilting its optical axis. The parametric amplifier is noncritically type I phase matched by controlling the temperature of the crystal to the accuracy of 0.1°C. After amplification the pump is eliminated by a combination of a mirror and filters (M2). An etalon (ET), an interference filter (IF), and an iris (I) of about 2 mm in diameter are placed to reduce the spurious fluorescence which has different directions or frequencies from those of the input signal. The remaining components are divided by a beam splitter (BS) and photon counting is performed by two silicon avalanche photodiodes (APD, RCA model SPCM-100-PQ, quantum efficiency at 1.064 μm~0.8%). Their outputs are fed to the start and stop inputs of a time-to-amplitude converter (TAC). The overall efficiency η of the detection was estimated to be 5×10^{-4} from measured optical losses and quantum efficiency of the detectors specified by a manufacturer. It is worth noting that the low detection efficiency does not affect the measured value of $g_n^{(2)}$ since in Eq. (1) both the numerator and the denominator are multiplied by η^2.

In Fig. 2 two experimental results obtained under the same condition except for the relative phase between the signal and the pump are shown. They represent the number of detected photon pairs as a function of the time delay. The data consist of peaks at 12 nsec ($= T$) intervals corresponding to the repetition rate of the mode-locked laser. The counts in the peak at time delay nT represent photon pairs detected in pulses nT apart. The width of the peaks is determined by the detector resolution time and is much larger than the pulse duration of the light. Hence, the total number of counts in a single peak rather than the height of the peak is relevant. In Fig. 2(a), the relative phase is chosen so that the output power of the OPA is minimum. As is seen in the figure, the peak at zero time delay is smaller than any other peak. This indicates the tendency that photon pairs are detected less frequently in a single pulse than in neighboring pulses, i.e., antibunching. Figure 2(b) displays the data when the relative phase is changed so that the output becomes maximum. It indicates the opposite effect, bunching.

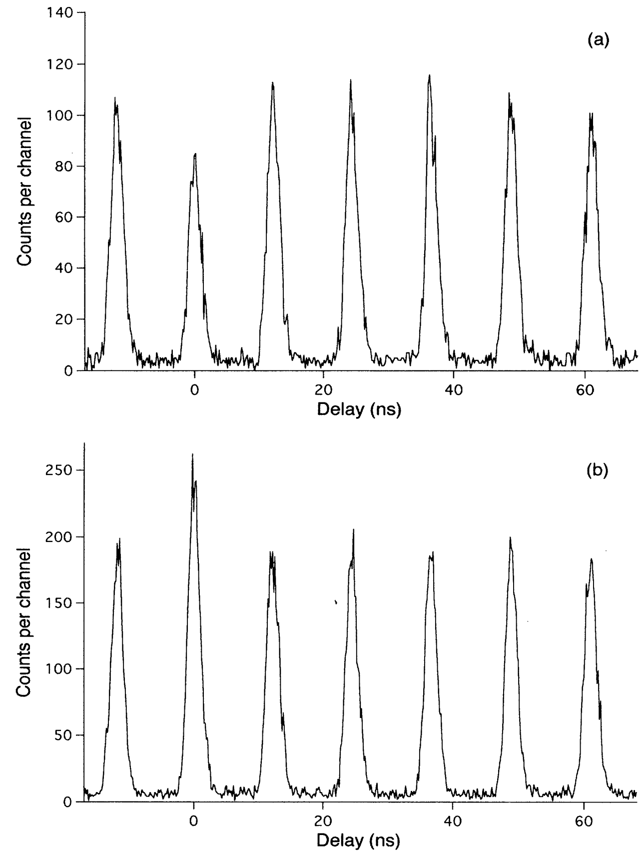

FIG. 2. The number of recorded photon pairs as a function of time delay. The channel width is 0.183 nsec. Input signal intensity is 0.28 photons per pulse, squeezing parameter $r = 0.084$, and the relative phase is adjusted (a) for maximum deamplification ($\varphi = 0$) and (b) for maximum amplification ($\varphi = \pi$) of the input field.

The normalized intensity correlation $g_n^{(2)}$ of the output light is calculated from the above data as follows. In the recorded correlation, scattered laser light and the dark counting of the detectors provide a background. When we take it into account, the sum of the counts $G_n^{(2)}$ recorded at time delays between $(n - 1/2)T$ and $(n + 1/2)T$ is related to $g_n^{(2)}$ by

$$G_n^{(2)} = (N_s T/\mathcal{R}_1)\{\mathcal{R}_1\mathcal{R}_2 + R_1 R_2 (g_n^{(2)} - 1)\}, \qquad (2)$$

where N_s is the number of the start pulses which actually activate the TAC, R_i and $\mathcal{R}_i$ $(i = 1, 2)$ are the counting rates at the detector i contributed by the output light alone and by both the output and the background, and the suffixes 1 and 2 refer to the start and stop channels [1,12]. We can measure the contribution of the background by blocking the path of the signal light: $\mathcal{R}_1 - R_1 = 1020$/sec and $\mathcal{R}_2 - R_2 = 810$/sec. N_s and $\mathcal{R}_i$ are directly measurable for each experiment. For Fig. 2(a), using $G_0^{(2)} = 1275$, $N_s = 1.103 \times 10^7$ for the measurement time of 1000 sec, $\mathcal{R}_1 = 11\,400$/sec, and $\mathcal{R}_2 = 11\,500$/sec, we obtain $g_0^{(2)} = 0.80$. The other six $g_n^{(2)}$ $(n = -1, 1, 2, \ldots, 5)$ are found to be almost unity: the average and the standard deviation of them are 1.003 ± 0.026. This deviation is consistent with the number of counts $G_n^{(2)} \sim 1.5 \times 10^3$. Thus we conclude that all $g_n^{(2)}$ $(n \neq 0)$ are unity and $g_0^{(2)}$ is smaller than those. This evidences that the light deamplified by the OPA shows antibunching ($g_0^{(2)} < g_n^{(2)}$), and the statistics of photon number in a pulse are sub-Poissonian ($g_0^{(2)} < 1$). For Fig. 2(b), $g_0^{(2)}$ is calculated to be 1.34 using $G_0^{(2)} = 3446$, $N_s = 1.436 \times 10^7$, $\mathcal{R}_1 = 14\,500$/sec, and $\mathcal{R}_2 = 15\,100$/sec. The other $g_n^{(2)}$ $(n \neq 0)$ are 1.015 ± 0.024 and can be similarly regarded as unity. This shows that the light amplified by the OPA has the bunching property and the super-Poissonian statistics. These results also show that two types of quadrature squeezing of coherent states, often referred to as amplitude squeezing and phase squeezing [13], are obtained via parametric amplification.

Figure 3 is a plot of $g_0^{(2)}$ for the two phases corresponding to the amplification and deamplification with various input signal intensities represented by the number of photons per pulse. The intensity of the pump is fixed for all data. Input intensities were estimated from the counting rates, the classical gain of the OPA, and

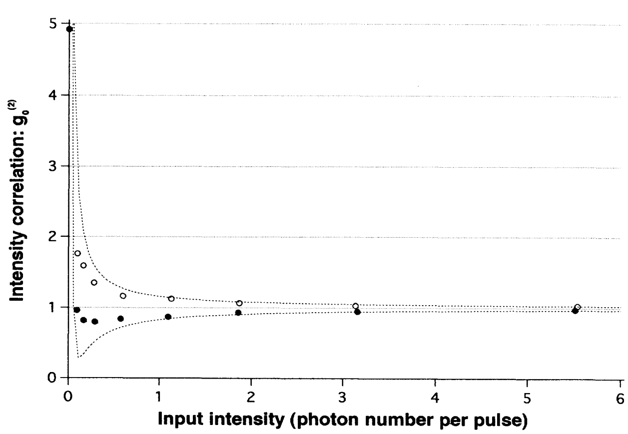

FIG. 3. Normalized intensity correlation $g_0^{(2)}$ for various input signal intensities. The relative phase φ is chosen for maximum deamplification (solid circles) and for maximum amplification (open circles). The data at zero input are independent of the relative phase. The dashed lines are the prediction by the theory using a single-mode picture.

the overall efficiency η. The classical gain was determined to be 1.18 from the two counting rates when the relative phase was set for amplification and for deamplification. We can see from Fig. 3 that the plotted $g_0^{(2)}$ shows appreciable deviation from unity when less than a few photons are expected in a pulse. $g_0^{(2)}$ for amplification rises monotonously from unity as the input intensity decreases, and reaches its maximum when there is no input and only parametric fluorescence exists. On the other hand, there is an optimum input intensity which minimizes $g_0^{(2)}$ for deamplification. As the input intensity decreases further, $g_0^{(2)}$ grows larger and finally reaches the same value as that for amplification.

The behavior of $g_0^{(2)}$ may be understood by a simple theory assuming a single mode. In this theory the pump is treated classically and losses are neglected [8]. Then, parametric amplification is expressed by the following relations,

$$b = a \cosh r - a^\dagger e^{-i\phi} \sinh r\ , \tag{3}$$

$$b^\dagger = a^\dagger \cosh r - a e^{i\phi} \sinh r\ , \tag{4}$$

where $a^\dagger$ and a are the creation and annihilation operators of the input field, $b^\dagger$ and b are those of the output field, r is a squeezing parameter, and ϕ the phase of the pump. When the input field is in a coherent state $|\alpha\rangle$, $g_0^{(2)}$ is evaluated as follows:

$$g_0^{(2)} - 1 = \frac{\langle\alpha|b^\dagger b^\dagger b b|\alpha\rangle}{\langle\alpha|b^\dagger b|\alpha\rangle^2} - 1$$

$$= \frac{A(r) + |\alpha|^2[B(r) - C(r)\cos\varphi]}{[(\cosh 2r - \sinh 2r \cos\varphi)|\alpha|^2 + \sinh^2 r]^2}\ , \tag{5}$$

where

$$A(r) = \cosh 2r \sinh^2 r\ , \tag{6}$$

$$B(r) = \cosh 4r - \cosh 2r\ , \tag{7}$$

$$C(r) = \sinh 4r - \sinh 2r\ , \tag{8}$$

$$\alpha = |\alpha| e^{-i\theta}\ , \tag{9}$$

$$\varphi = 2\theta - \phi\ . \tag{10}$$

In the above expression, φ represents the relative phase between the input and the pump. The output intensity $\langle\alpha|b^\dagger b|\alpha\rangle$ is maximum for $\varphi = \pi$ and minimum for $\varphi = 0$. The first term of the numerator in Eq. (5), $A(r)$, does not vanish even when $|\alpha|^2 = 0$, so it represents correlation of parametric fluorescence alone. It is always positive reflecting that an OPA emits two photons simultaneously as observed for nondegenerate [14] and degenerate [15] configurations. When $|\alpha|^2$ is very small, this positive term is dominant and $g_0^{(2)} - 1$ is always positive irrespective of the relative phase φ. When $|\alpha|^2$ is large enough to make the second term of the numerator dominant, the sign of $g_0^{(2)} - 1$ depends on φ. Production of signal photons from pump photons ($\varphi = \pi$, amplification) gives $g_0^{(2)} - 1$ a positive value, and removal of signal photons ($\varphi = 0$, deamplification) makes $g_0^{(2)} - 1$ negative [note that $C(r) > B(r) > 0$]. In the case where $|\alpha|^2$ largely exceeds unity, $g_0^{(2)} - 1$ approaches zero with $|\alpha|^{-2}$ or, in other words, with $\hbar\omega/E$ where E is energy per pulse. The deviation of $g_0^{(2)}$ from unity in Eq. (5) originates from the commutation relation of operators. It is therefore a quantum mechanical effect and disappears when the energy becomes macroscopic.

For the comparison with the theory, the squeezing parameter r in the experiment for Fig. 3 was determined to be 0.084 by equating the measured classical gain, 1.18, with e^{2r}. Using this value, the theoretical prediction of Eq. (5) for $\varphi = 0$ and π is represented by the dashed lines in Fig. 3. The discrepancy between the measured data and the single-mode theory for low input intensities is mainly due to spontaneous fluorescence which is generated at nondegenerate frequencies and passes through the etalon and the iris. Given r and η, we can estimate how much fluorescence should be detected from the single-mode theory described above. The measured counting rate when there is no input is, however, about 6 times larger than the estimation. This suggests that the fluorescence in unseeded modes also reaches the detectors. Such fluorescence may cause the effect similar to stray light and make the measured $g_0^{(2)}$ at weak input intensities closer to unity than the dashed lines, as can be seen in Fig. 3. In order to observe more prominent antibunching, it would be important to reduce such fluorescence effectively.

Mandel's Q parameter [16] is often used for sub-Poissonian statistics. For comparison, we can calculate it as follows:

$$Q \equiv \frac{\langle(\Delta n)^2\rangle - \langle n\rangle}{\langle n\rangle} = R_1 T(g_0^{(2)} - 1). \qquad (11)$$

For Fig. 2(a), we obtain $Q = -3 \times 10^{-5}$. This value is smaller than those in Refs. [2,4,5,17] because of the smaller quantum efficiency of the detectors at 1.06 μm. If this is not the case, the Q value can be improved since the output light is collimated well compared to the fluorescence experiments.

In summary, we have investigated the intensity correlation of pulsed squeezed light generated by a degenerate OPA pumped by pulsed light. We used very weak pulsed coherent light as the input signal: less than several photons in a pulse on the average. The measured correlation showed bunching when the relative phase between the signal and the pump was adjusted for amplification. When we chose the phase for deamplification, the data revealed the opposite phenomenon, antibunching, and sub-Poissonian statistics, which are nonclassical properties of light.

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