Bunching of Bell states

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The bunching of two single photons on a beam-splitter is a fundamental quantum effect, first observed by Hong, Ou and Mandel. It is a unique interference effect that relies only on the photons’ indistinguishability and not on their relative phase. We generalize this effect by demonstrating the bunching of two Bell states, created in two passes of a nonlinear crystal, each composed of two photons. When the two Bell states are indistinguishable, phase insensitive destructive interference prevents the outcome of four-fold coincidence between the four spatial-polarization modes. For certain combinations of the two Bell states, we demonstrate the opposite effect of anti-bunching. We relate this result to the number of distinguishable modes in parametric down-conversion.

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The bunching of photons is a manifestation of their bosonic nature. Although resulting from quantum interference, it is insensitive to the photons’ relative phase. Bunching of two photons on a beam-splitter was first demonstrated in the famous experiment by Hong, Ou and Mandel (HOM)1. Later, bunched photon states were shown to be useful for quantum limited interferometric measurements2 and for beating the classical diffraction limit3. The bunching of three photons was also experimentally achieved4. Photon bunching became a fundamental tool in quantum optics. It is used in Bell state analysis5, teleportation6 and more. Recently, the basic principles of bunching have also been found to be useful for quantum information processing with linear optics7. An example is the nonlinear phase-shift on a beam-splitter for the quantum controlled-NOT gate8.

Photons can bunch also in circumstances where there are no beam-splitters. Using the equivalence between the operation of beam-splitters on two spatial modes and the operation of waveplates on two polarization modes9, a horizontally polarized and a vertically polarized photon can bunch when their polarization basis is rotated. Polarization interference experiments are simpler as they involve a smaller number of spatial modes.

The most common source for photon bunching experiments, as well as for photon entanglement, is parametric down-conversion (PDC). As an intense pump beam is passing through a crystal possessing χ\(^{(2)}\) nonlinearity, some of its photons can split into two. For type-II non-collinear PDC, these two photons are emitted into two spatial modes (referred to as modes a and b), and can exhibit polarization entanglement10. It is helpful to use pulsed pump sources due to their well defined timing. Recently, many experiments have used configurations where a pump pulse passes through the nonlinear crystal twice11. In this Letter we show how four photons that originate from such two passes avoid being equally distributed between the four possible spatial-polarization modes by phase-insensitive destructive interference12. This result has many similarities to the HOM bunching.

Its main difference though is that instead of the bunching of two single photons in Fock states, the bunching occurs between two composite states – the Bell states. As opposed to previous double-pass experiments, the result is unaffected by the amplitudes of two-pair emission in one of the two passes. We also show how, unlike in the HOM case of single photon states, for certain combinations of Bell states the bunching transforms into anti-bunching.

Non-collinear type-II parametric down-conversion creates the following bi-partite state in a single pass of the nonlinear crystal13

\[
|\psi\rangle = \frac{1}{\cosh^2 \tau} \sum_{n=0}^{\infty} \sqrt{n+1} \tanh^n \tau |\psi_n^-\rangle, \quad (1a)
\]

\[
|\psi_n^-\rangle = \frac{1}{\sqrt{n+1}} \sum_{m=0}^{n} (-1)^m |n-m, m\rangle_a|m, n-m\rangle_b. \quad (1b)
\]

where \(|n, m\rangle\_a\) represents \(m\) horizontally and \(n\) vertically polarized photons in mode \(i\). The magnitude of the interaction parameter \(\tau\) depends on the nonlinear coefficient of the crystal, its length and the intensity of the pump pulse. The one-pair term \((n=1)\) is the familiar \(\psi^-\) Bell state14. We concentrate on the case when two indistinguishable photon pairs are produced \((n=2)\). This term contains three elements and it is written in the above formalism as

\[
|\psi^-_2\rangle = \frac{1}{\sqrt{3}} (|2, 0\rangle_a|0, 2\rangle_b - |1, 1\rangle_a|1, 1\rangle_b + |0, 2\rangle_a|2, 0\rangle_b). \quad (2)
\]

Previously, it has been shown how a four-fold coincidence between the four modes \(a_h, a_v, b_h\) and \(b_v\) is forbidden for the state of Eq. 2 when the photons of mode \(a\) are rotated to an orthogonal polarization basis compared to the photons of mode \(b\)15, 16, 17. For example, if mode \(a\) is observed at the linear horizontal-vertical (hv) basis and mode \(b\) at the linear plus-minus (pm) 45° basis (or the right-left circular basis \(rl\)), an \(a_h a_v\) coincidence collapses the state to the middle term of Eq. 2 and the polarization rotation bunches the \(b\) mode photons such
that $b_h b_v \sim b_p^2 - b_m^2$. Thus, a four-fold coincidence of the form $a_h a_v b_p b_m$ can never occur. The same result applies to any two orthogonal polarization bases due to the rotational symmetry of Eq. 2. Nevertheless, when more than one mode are collected for each spatial-polarization mode (as in a double-pass configuration), these four-fold coincidences can be observed.

In the experiment, we used a PDC source in a double-pass configuration. The nonlinear crystal is pumped by 200 fs pulses of 390 nm wavelength from a doubled Ti:Sapphire laser at a repetition rate of 80 MHz. Down-converted photons from the first pass are redirected into the crystal to meet again with the same pump pulse. The down-converted photons from the second pass are re-injected into the crystal together with the pump pulse. The temporal walk-off. Photons are coupled through narrow band-pass filters into single-mode fibers and detected by APDs.

For a pure $\psi_2^-$ state, no four-fold events can be detected at orthogonal polarization bases and zero delay. Nevertheless, when $\Delta t \gg t_c$, the temporal delay adds distinguishability between the Bell states from the first and the second passes, doubles the number of collected modes and gives rise to four-fold coincidences. To demonstrate this, we define a ladder operator $L^t = (a_h^\dagger b_v^\dagger - a_v^\dagger b_h^\dagger)/\sqrt{2}$, composed of creation operators of the four PDC modes. The operator $L^t$ creates a $\psi_1^-$ state when applied once to the vacuum and a $\psi_2^-$ state when applied twice. The four-photon state resulting from two passes of a pump pulse in the down-conversion crystal with a phase difference of $\omega \Delta t$ between them is 

$$|\psi_2\rangle = \frac{1}{4}(L^t_1 + e^{i\omega \Delta t} L^\dagger_1)|\text{vac}\rangle$$

$$= \frac{1}{4}(L_1^2 + 2e^{i\omega \Delta t} L_1^\dagger L_1^\dagger + e^{2i\omega \Delta t} L_1^\dagger L_1^\dagger)|\text{vac}\rangle.$$  

The roman digits designate a distinguishing quantum number. The distinction can be one of many options, such as the photon wavelength, spatial mode or timing, as in this double-pass case. The terms that result from the first and last operators in Eq. 3 do not contribute to the $a_h a_v b_p b_m$ four-fold coincidence as they each create a $\psi_2^-$ state. Keeping the pass number labels and discarding normalization, the resulting evenly populated state terms, that would yield a four-fold event when observing modes $a$ and $b$ at orthogonal polarization bases are

$$|\psi_{4-fold}\rangle = (a_h^\dagger a_v^\dagger b_p^\dagger b_m^\dagger + a_h^\dagger a_v^\dagger b_p^\dagger b_m^\dagger - a_h^\dagger a_v^\dagger b_p^\dagger b_m^\dagger)|\text{vac}\rangle.$$  

Because these terms originate only from the middle term $L_1^t L_1^\dagger$ of Eq. 3 their amplitudes are neither sensitive to the phase $\omega \Delta t$, nor to the amplitude balance between the two passes. Like in the HOM bunching experiment, when the two input states are indistinguishable, the pass indices are omitted and the amplitude for $\psi_{4-fold}$ disappears. However, the absence of interference in the distinguishable case revives the four-fold amplitude. As the delay $\Delta t$ is scanned the distinguishability is varied, resulting in a dip for the four-fold counts, centered at zero delay.  

Figure 2 shows the four-fold coincidence rate for orthogonal polarization bases as a function of the delay $\Delta t$ between two $\psi^-$ states. Mode $a$ is detected at the $hv$ basis and mode $b$ is at $pm$. Every point is integrated over 25 minutes. The curve for the best fit value of $\alpha = 0.8$ is solid while predictions for other $\alpha$ values are shown as dashed lines. Inset: a fine scan around zero delay reveals oscillations.
between two $\psi_1^-$ states. As predicted, the coincidence rate has a dip, centered at the zero delay point. The dip width corresponds to the coherence time of the two $\psi_1^-$ states.

Previously, the quality of the two-pairs state was defined by $\alpha$, the probability for having the $\psi_1^-$ state, as opposed to two distinguishable $\psi_1^-$ states:  

$$|\psi\rangle = \sqrt{\alpha}|\psi_2^+\rangle + \sqrt{1-\alpha}|\psi_1^-\rangle \otimes |\psi_1^-\rangle.$$  

This model is problematic if we assume that it originated from two modes as in Eq. [8]. First, such a state has at least 60% content of $\psi_2^+$, i.e. $\alpha$ has a non-zero minimal value. Second, the $\psi_2^+$ content is the sum of two terms, $\psi_{2,1}$ and $\psi_{2,11}$ from the two modes (passes). Furthermore, two or more distinguishable modes are possible even for a single pass. We would like to have a model with arbitrary number of modes that will support results where $\alpha$ has lower values and explain the experimental imperfect interference at the center of the dip. The multi-mode Hamiltonian and the four-photon component of the state produced are

$$H = \frac{ik}{n_d} \sum_{j=1}^{n_d} c_j \epsilon^{i\theta_j} L_j^\dagger + h.c.,$$  

|\phi_2(n_d)| = \frac{1}{\sqrt{C}} \left( \frac{1}{n_d} \sum_{j=1}^{n_d} c_j \epsilon^{i\theta_j} L_j^\dagger \right)^2 |\text{vac}\rangle,  

where $\kappa$ is a coupling constant that depends on the non-linearity of the crystal and the intensity of the pump pulse, and $C$ is the proper four-photon component normalization. The distribution of $c_j$ (real numbers) determine the number of modes involved and their relative weight such that their average is $\langle c \rangle = 1$. As the operator $L_j^\dagger$ has two terms, the state of Eq. [8] has $2n_d^2 + n_d$ non-interfering terms ($3n_d$ originate from $(L_j^\dagger)^2$ terms and $2n_d(n_d - 1)$ from $L_j^\dagger L_k^\dagger$), of which only $n_d^2 - n_d$ can give rise to the relevant four-fold events (half of the $L_j^\dagger L_k^\dagger$ terms). For the simple case of equally weighted modes, the intensity of $|\phi_2\rangle$ is $(2n_d + 1)/(n_d^3)$, which is higher for smaller $n_d$ values due to better stimulation. After rotation to orthogonal polarization bases, four-fold events result only half the time and for the equally weighted modes case their probability per pump pulse scales as

$$P_3(n_d) \propto \frac{n_d - 1}{2n_d^2}.  

When only one PDC mode is collected there are no such events, and when the mode count increases, the rate peaks for $n_d = 2$ and decays for larger numbers. When the time delay $\Delta t$ is introduced in the experiment, the mode number $n_d$ is doubled such that each term from the first pass corresponds to a delayed term from the second pass.

$$H = \frac{1}{2}(H_1 + e^{i\omega\Delta t} H_{11})$$  

At large delays $\Delta t \gg t_c$ the two corresponding modes are distinguishable and Eq. [8] still holds with $n_d$ replaced by $2n_d$. This case is equivalent to doubling $t_c$ (see Ref. [19]). When the delay is not large enough for distinguishability, the two passes of Eq. [9] interfere and create oscillations between zero and an upper bound defined by Eq. [8]. Therefore, in the single-mode case, a scan will change the mode number between 2 and 1, thus the four-fold event count would present a phase-independent dip. When more modes are collected, the rate would oscillate between zero and a peak envelope. The insert in Fig. 2 is a fine scan that reveals the expected oscillations.

Equation [8] is a parameterization of Eq. [5] for the case of $n_d = 2$ and different weights, where $\alpha = \frac{1}{1+\sqrt{2}}(c_1^2 + c_2^2)$. Thus, the four-fold rate result of Fig. 2 is composed of a dip contribution from a single mode per pass element and an oscillating contribution from a two modes per pass element. Because the dip result of Fig. 2 was integrated over a long time, phase fluctuations from experimental instability averaged the oscillating term. Considering this model with the averaging effect, the best fit for the dip data corresponds to $\alpha = 0.80 \pm 0.05$. The dashed curves in Fig. 2 demonstrate the predictions for different $\alpha$ values. The dip is sensitive to changes in $\alpha$ and transforms into a peak for $\alpha \lesssim 0.64$.

Great care has to be taken to ensure that the collected photons originated from a single mode. Such measures include wavelength and spatial filtering and compensation for temporal and spatial birefringence walk-offs. Chirped pump pulses are also a source for distinguishability as their duration is longer than their coherence length. Originally, it was suggested to measure $\alpha$ through the four-fold visibility, i.e. the contrast between four-fold coincidence count rates at similar and orthogonal polarization bases. Previously published values of $\alpha$ are 37% and 83%, in Refs. [18] and [19], respectively. The results of the four-fold visibility of our setup are presented in Fig. 2. From the data we calculated $\alpha = 86\%$. The agreement with the fit value of $\alpha = 80 \pm 5\%$ from
in a specific setup. The sensitive bunching measurement is efficient for obtaining the number of PDC modes collected. Bell state bunching effects are insensitive to the amplitudes of two pairs from one pass (first and last terms of Eq. 3), unlike any previous experiment. It was shown how the bunching arises from varying the phase delay as before. The bunching contrast was related to the ψ2 content of the four photon state. Just like the HOM effect plays a crucial role in projective measurements and operations between single photons, the bunching of Bell states might prove to be a useful tool in manipulating and projecting multi-photon states.

Table I: Bunching and anti-bunching of Bell states for two polarization options of the modes (a, b). The letters B and A mark bunching and anti-bunching combinations, respectively.

| (hv, pm) | ψ− | ψ+ | φ− | φ+ |
|---------|-----|-----|-----|-----|
| ψ−      | B   | B   | B   | A   |
| ψ+      | B   | B   | A   | B   |
| φ−      | B   | A   | B   | B   |
| φ+      | A   | B   | B   | B   |

FIG. 4: Four-fold coincidence rate as a function of the delay Δt between a ψ− and a φ+ states. Polarization bases and all other parameters are the same as in Fig. 2. Solid line is a Gaussian fit. Anti-bunching by a factor of 2.6 is observed.

In conclusion, we presented the bunching and anti-bunching of two pairs of photons, each in a Bell state. Only the amplitudes of a single pair from each of the two crystal passes of a pump pulse contribute to this effect. It was shown how the bunching arises from varying the number of distinguishable modes by adding a time delay between the two pairs. The bunching contrast was related to the ψ2− content of the four photon state. Just like the HOM effect plays a crucial role in projective measurements and operations between single photons, the bunching of Bell states might prove to be a useful tool in manipulating and projecting multi-photon states.

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