Observation of Four-Photon Interference with a Beam Splitter by Pulsed Parametric Down-Conversion

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When four photons arrive at a beam splitter, two from each side, a four-photon, six-path interference effect occurs to yield a sixfold enhancement of the probability for all four photons to exit together from the beam splitter. We produce the four-photon state by using the stimulated emission process in a pulsed parametric down-conversion and measure the probability for all four photons to exit from one side of the beam splitter. The observed enhancement factor is in good agreement with a multimode treatment of pulsed down-conversion.

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The generation of multiparticle entangled quantum states has attracted much attention in recent years for their potential applications in communication, computation [1], and more accurate atomic frequency standards [2]. Such quantum states with three or more particles can also display dramatic locality violation as discovered by Greenberger, Horne, and Zeilinger (GHZ) [3]. Schemes have since been proposed to produce such states based on the superposition of independent pairs of photons generated from parametric down-conversion [4,5]. Although four-photon coincidences were measured in experiments on quantum state teleportation [6] and swapping [7], involving two independent parametric down-converters, the underlying principle is still two-photon interference. On the other hand, all of the higher-order interference schemes so far (including the GHZ multiparticle interferometry) involve only the quantum interference between two alternative paths and usually result in sinusoidal modulations in the coincidences. When more alternative paths become involved, however, new and interesting phenomena arise. For example, Shor's factorization algorithm [8] of quantum computation relies on a multiple path quantum interference effect to achieve massive parallelism. A similar effect also occurs in optical gratings. Furthermore, complete constructive or destructive quantum interference (i.e., with 100% visibility) is important for such an algorithm to yield the correct outcome with a high degree of certainty although quantum processes usually are associated with randomness.

In this Letter, we report a four-photon multiple-path (six) interference effect that manifests in the partition ratio for the four photons when they arrive at a 50:50 beam splitter, two from each side. The four-photon quantum state is produced by using the stimulated emission process in a pulsed parametric down-converter. We observe a more than fivefold (theoretically, sixfold) increase in the quadruple coincidence at one exit port of the beam splitter when interference occurs. Such an increase can only be explained by the four-photon interference.

It is well known by now that, when two identical photons arrive at a 50:50 beam splitter, one from each side, a two-photon interference effect occurs such that the two photons will exit together from the same side of the beam splitter [9]. This type of two-photon interference effect played an important role in the studies of nonlocality [10] and quantum interference [11,12]. When four photons enter a beam splitter, two from each side as shown in Fig. 1, what are the partition probabilities for the four photons between the two exit ports? One may be tempted to consider the four photons as two pairs of photons and predict the outcome by using the known two-photon effect. This is true when the four photons are made of two pairs of photons independent and identifiable from each other. However, when the four photons are identical and therefore unidentifiable, a four-photon interference effect occurs and the outcome is quite different from that of the two-photon situation.

It can be shown [13] that for an input state of $|2,2\rangle$ at a 50:50 beam splitter, the output state becomes

$$|\Psi\rangle = \sqrt{\tfrac{3}{8}}\,(|4,0\rangle + |0,4\rangle) + \tfrac{1}{2}\,|2,2\rangle\,. \tag{1}$$

The coefficient of $\sqrt{3/8}$ can be easily obtained by using two methods to calculate the four-photon probability $\langle \hat{A}^{\dagger 4}\hat{A}^{4}\rangle$ and require them to be equal: The first method uses directly the components (e.g., $|4,0\rangle$) in the output

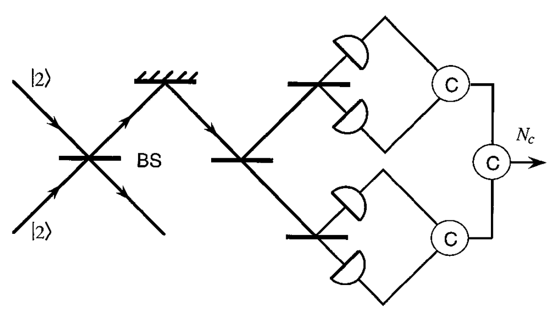

FIG. 1. Schematic illustration of the four-photon interference and partition at a beam splitter.

state in Eq. (1); the second method relates the beam splitter's output operator $\hat{A}$ with its input operators $\hat{a}$ and $\hat{b}$ by $\hat{A} = (\hat{a} + i\hat{b})/\sqrt{2}$ to carry out the evaluation. The result in Eq. (1) is completely different from the predictions if one were to treat the four incoming photons as classical particles where a simple Bernoulli probability distribution applies. The classical probability for $|4,0\rangle$ would simply be $(1/2)^4 = 1/16$, as compared to 3/8 in Eq. (1). The classical probability for $|3,1\rangle$ or $|1,3\rangle$ is a nonzero value of 1/4. But the $|3,1\rangle$ and $|1,3\rangle$ states are absent in the output state in Eq. (1). The classical probability for a $|2,2\rangle$ state is 3/8. But Eq. (1) predicts it to be 1/4. Although the disappearance of the $|3,1\rangle$ and $|1,3\rangle$ terms in Eq. (1) can be explained by the two-photon interference [known as "2×2," see below in Eq. (2)], the sixfold increase in the probabilities for $|4,0\rangle$ and $|0,4\rangle$ can be explained only by the quantum interference of the four input photons. If it were due to two-photon interference (2×2), the effect on the probability distribution would come from the cases where the two pairs are independent of each other. In this case, the two independent pairs of twin photons will interfere individually and produce the state [9]

$$|\Psi\rangle = |\psi\rangle \times |\psi\rangle, \quad \text{with } |\psi\rangle = \frac{1}{\sqrt{2}}(|2,0\rangle + |0,2\rangle). \tag{2}$$

This will give rise to a probability of 1/4 for $|4,0\rangle$, a fourfold rather than sixfold increase when compared to the classical prediction. On the other hand, a classical wave theory predicts only an enhancement of 4.375-fold for a four-beam joint intensity measurement and it cannot explain the missing $|3,1\rangle$ term in Eq. (1).

In order to understand the sixfold increase in quadruple coincidence in terms of the four-photon interference, we consider the events in which each of the four detectors detects a photon and results in a quadruple coincidence event (Fig. 1). We notice that there are six possible ways ($4!/2!\,2! = 6$), with an equal amplitude χ, to arrange the four identical photons' exit from the beam splitter among the four detectors. The division by the factor of $2!\,2!$ comes from the fact that each input beam of the beam splitter contains two identical photons. The six probability amplitudes interfere constructively in the four-photon detection probability, resulting in an overall probability of $(6\chi)^2 = 36\chi^2$ for the quadruple detection. But, without interference, the six probabilities (χ^2) simply add to give a $6\chi^2$ overall quadruple probability in a classical case. Thus we will have a sixfold increase due to quantum interference as compared to a classical prediction. As seen from the argument above, a multiple-path (six) four-photon interference is responsible for the sixfold increase. Similarly, for the overall probability of the output state $|2,2\rangle$, there are also six possibilities. But because of the π-phase shift for reflected photons, two of the six probability amplitudes interfere with the other four destructively, resulting in a probability of $(2\chi)^2 = 4\chi^2$, or a reduction in the partition probability by a factor $(4\chi^2/6\chi^2) = 2/3$.

It is well known that a two-photon state can be generated via spontaneous parametric down-conversion. To produce a four-photon state, however, we must consider the stimulated emission process. The extra two photons are induced by the original two spontaneously emitted photons. This involves a higher-order process. To see this more clearly, we consider the Hamiltonian for the parametric down-conversion process:

$$\mathcal{H} = \kappa \hat{a}_s^\dagger \hat{a}_i^\dagger + \text{H.c.} \tag{3}$$

The state of the down-converted fields becomes

$$\begin{aligned} |\Psi\rangle &= \exp(-i\mathcal{H}t/\hbar)|vac\rangle \\ &\approx [1 - i\mathcal{H}t/\hbar + (-i\mathcal{H}t/\hbar)^2/2]|vac\rangle, \end{aligned} \tag{4}$$

where we have dropped the higher-order terms in this approximation. We can rewrite the quantum state as

$$|\Psi\rangle = (1 - \eta^2/2)|vac\rangle - i\eta|1_s, 1_i\rangle - \eta^2|2_s, 2_i\rangle, \tag{5}$$

where $\eta \equiv \kappa t/\hbar$ is a dimensionless coupling constant. The state is normalized up to the order of η. Obviously, the second term in Eq. (5) gives a two-photon state while the last term corresponds to a four-photon state with two in each of the signal and idler modes, respectively. Using a quadruple coincidence detection scheme (Fig. 1), the first two terms in Eq. (5) do not contribute and we thus detect only the required $|2,2\rangle$ state.

The four-photon term in Eq. (5) can be viewed as a result of the stimulated emission (amplification) induced by the two-photon term. In fact, parametric down-conversion produces thermal fields for each of the down-converted beams [14]. Therefore, the four-photon term is related to the photon bunching effect or the Hanbury Brown-Twiss effect [15]. Indeed, a simple calculation shows that the normalized intensity correlation function $g_s^{(2)} = g_i^{(2)} = 2$ for the state in Eq. (5) [16], which is exactly what we expect for a single mode thermal field. However, the process of parametric down-conversion has a very wide spectrum. When we apply a multimode treatment for down-conversion, we find that $g_s^{(2)}(\tau) = 1 + |\gamma(\tau)|^2$ which depends on the time delay τ between the detections of two photons in one beam. $\gamma(\tau)$ is the normalized second-order correlation function and has the properties that $|\gamma(\tau)| \leq 1$ and $\gamma(\infty) = 0$. This comes from stimulated emission and gives rise to photon bunching. The width of $\gamma(\tau)$ is of the order of the reciprocal bandwidth for the field. Thus in order to observe the four-photon contribution in Eq. (5), one must use detectors which have a response time faster than the fluctuations of the fields. Unfortunately, with the wide bandwidth of down-conversion, no optical detector resolves it. As an alternative solution, one can use ultrafast pulses to pump the down-converters [17,18] and use a narrow filter to create a transform-limited down-conversion pulses [19]. Such a strategy has also been applied in recent experiments on teleportation and quantum state swapping [6,7].

When pumped by pulses, the down-converted fields are generated in the form of wave packets. For short wave packets, slow optical detection results in an integration over the period of the entire pulse. We developed a multimode theory for pulsed parametric down-conversion [20] that gives rise to the two-photon coincidence probability for a single down-conversion beam as

$$P_2 = \mathcal{A} + \mathcal{E}\,. \tag{6}$$

Here, $\mathcal{A}$ is the accidental coincidence and $\mathcal{E}$ is the in-excess coincidence resulting from the photon bunching effect [$\mathcal{E} \le \mathcal{A}$ because $|\gamma(\tau)| \le 1$].

Now we consider the case when both beams of down-conversion are superposed at a beam splitter as in Fig. 2. We examine the probabilities for all four photons to exit together from one side of the beam splitter. When the two wave packets do not overlap at the beam splitter, or the delay between the arrival times of the two down-converted wave packets is much larger than their pulse widths, no interference effect occurs. We find that the four-photon coincidence for this case is given by

$$P_4(\infty) = \alpha(\mathcal{A} + \mathcal{E})\,, \tag{7}$$

where α is the four-photon collection efficiency. This corresponds to the classical case as discussed earlier and is similar to the one-beam case in Eq. (6). Conversely, when the two wave packets overlap at the beam splitter or, equivalently, the delay is 0, we have, for the four-photon coincidence [20],

$$P_4(0) = 4\alpha(\mathcal{A} + 2\mathcal{E})\,. \tag{8}$$

Here the factor 4 comes directly from the 2×2 case while the factor 2 in front of $\mathcal{E}$ is a result of four-photon interference. Thus the overall enhancement factor becomes

$$\frac{P_4(0)}{P_4(\infty)} = 4\left(1 + \frac{\mathcal{E}}{\mathcal{A} + \mathcal{E}}\right) \le 6\,. \tag{9}$$

The last inequality comes from the fact that $\mathcal{E} \le \mathcal{A}$. In the ideal case when $\mathcal{E} = \mathcal{A}$, the ratio is exactly 6, corresponding to the sixfold increase in the probability for $|4,0\rangle$ from the classical prediction to the quantum prediction as in Eq. (1). Therefore the multimode prediction covers the single mode case. Notice that, when $\mathcal{E} = 0$, we have the ratio equal to 4. This is the 2×2 case, where only two-photon interference contributes. In fact, when $\mathcal{E} = 0$, there is no stimulated emission and the only chance to create four photons is via an accidental production of two pairs of spontaneously emitted photons during a single pump pulse. On the other hand, it appears from Eq. (8) that the contribution from stimulated emission ($\mathcal{E}$ term) gives rise to an eightfold rather than sixfold increase as mentioned earlier. In fact, the stimulated emission photons cannot be separated from the spontaneous emission of the accidental pairs. Therefore, the simple picture of four-photon interference applies generally to any four single mode photons irrespective of whether they are independent or correlated, as long as they are identical. Thus the sixfold increase is the overall contribution from both spontaneous and stimulated photons.

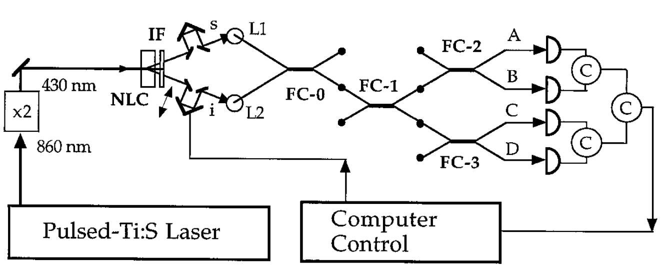

FIG. 2. Experimental setup. L1 and L2 are two lens systems that couple the signal and idler beams into the fiber system with high efficiencies.

The outline of the experimental setup is shown in Fig. 2. A Ti:sapphire mode-locked laser [Spectra Physics 3600D] produces 860 nm, transform-limited 150 fs pulses at a repetition rate of 80 MHz. The laser pulses are frequency doubled with a 1-mm-long $KNbO_3$ nonlinear crystal, and the resulting 430 nm pulse train is used to pump an identical nonlinear crystal (NLC) serving as a parametric down-converter. We tune the temperature of the down-converter such that the signal and idler beams at 860 nm are emitted at approximately 4° apart. After going through a 0.9 nm bandwidth interference filter (IF), the signal and idler beams are carefully aligned into the input ports of a 50:50 2×2 optical fiber coupler (FC-0, a fiber beam splitter). The polarization of one arm is adjusted by using a quarter-wave and a half-wave plate to obtain a maximum interference in the output ports of the 2×2 coupler. The relative time delay between the signal and idler photon wave packets is scanned by a computer-controlled translation stage.

First, the two output ports from the fiber coupler FC–0 are fed into two avalanche photodiode single photon detectors [EG&G SPCM-132] and the two-photon coincidence counts are measured as a function of the relative time delay. A coincidence dip of 90% modulation is observed [9]. Second, one of the two output ports of FC-0 is further split into four with fiber couplers FC-1, 2, and 3 as shown in Fig. 2. The four outputs are fed into four single photon detectors for quadruple coincidence measurement. Here, the quadruple coincidence events are the results of two possibilities. First, accidentally, two sets of signal-idler photon pairs are emitted from NLC during one pump pulse and, with a properly combined binary branching at all four fiber couplers, the four detected photons produce a quadruple coincidence count. This corresponds to an "accidental" coincidence event between the randomly produced photon pairs. As the

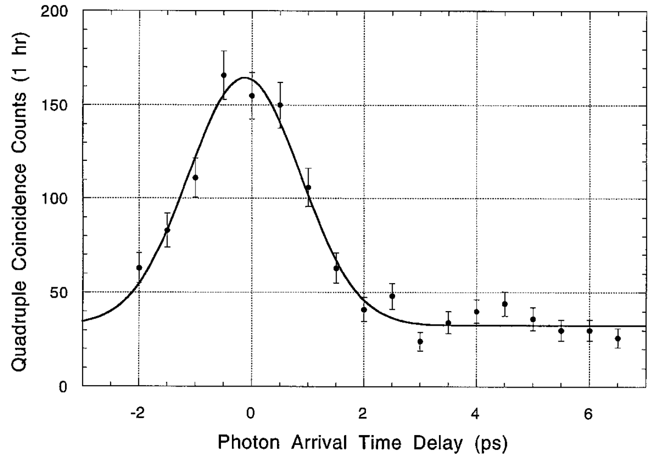

FIG. 3. Quadruple coincidence event versus relative time delay between the signal and idler photon wave packets. The counting time for each data point is one hour. The solid line is the result from a least-squares fitting procedure. The error bars indicate one standard deviation. The width of the curve is approximately the inverse of the 0.9 nm bandwidth of the filter IF.

second possibility, a pair of signal and idler photons is first generated in NLC and then amplified to produce a second identical pair. This creates an "in-excess" coincidence event. The relative ratio of the possibilities for the two events manifests in the form of the ratio $\mathcal{E}/\mathcal{A}$ as given in Eq. (7) and can be measured in an ancillary two-photon correlation experiment on one beam. For this purpose, we block off either the idler or the signal beam, and measure the coincidence rate between two detectors A and B. The resulting coincidence rate follows a simple relation $R_{AB} = (1 + \mathcal{E}/\mathcal{A}) \times R_A R_B / R_L$, where R_{AB}, R_A, R_B, and R_L are the counting rates for coincidence between A and B, for detectors A and B, and the 80 MHz laser repetition rate, respectively. Ratios of $\mathcal{E}/\mathcal{A} = 0.7$ and 0.6 are measured for the signal and idler beams, respectively. The different values of $\mathcal{E}/\mathcal{A}$ for the signal and idler beams are caused by slight misalignment at the fiber couplers and the imperfection of spectral filtering resulting in a slight difference in the collection of spectral components.

With all beams exposed, the quadruple coincidences are then measured as a function of the relative time delay between the signal and idler beam paths using the configuration shown in Fig. 2. The result is shown in Fig. 3. A least-squares fitting procedure is employed and gives a ratio of $R_4(0)/R_4(\infty) = 5.09 \pm 0.37$. This is lower than the prediction of $R_4(0)/R_4(\infty) = 5.58$ according to Eq. (9) when we use an average value of $\mathcal{E}/\mathcal{A} = 0.65$. However, a more complete analysis [20] that takes into account the less than perfect alignment (results in an observed 90% visibility for the two-photon coincidence dip) yields a prediction of $R_4(0)/R_4(\infty) = 5.06$, in good agreement with the experiment.

In conclusion, we analyzed the interesting situation of the partition at a beam splitter for four photons generated by a parametric down-converter using pulsed pumping. We point out the different partition ratios for identifiable [(2×2)-photon] and unidentifiable (four-photon) pairs of photons. The latter is a result of stimulated emission in parametric down-conversion. We demonstrated for the first time such a multiparticle, multiple path, interference effect using a pulse-pumped parametric down-conversion.

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[1] A. Steane, Rep. Prog. Phys. **61**, 117 (1998).
[2] J. J. Bollinger *et al.,* Phys. Rev. A **54**, R4649 (1996).
[3] D. M. Greenberger, M. A. Horne, and A. Zeilinger, in *Bell's Theorem, Quantum Theory, and Conceptions of the Universe,* edited by M. Katafos (Kluwer, Dordrecht, 1989).
[4] B. Yurke and D. Stoler, Phys. Rev. Lett. **68**, 1251 (1992).
[5] A. Zeilinger *et al.,* Phys. Rev. Lett. **78**, 3031 (1997).
[6] D. Bouwmeester *et al.,* Nature (London) **390**, 575 (1997).
[7] J. W. Pan *et al.,* Phys. Rev. Lett. **80**, 3891 (1998).
[8] P. Shor, in *Proceedings of the 35th Symposium on Foundations of Computer Science* (IEEE Comp. Soc. Press, Los Alomitos, CA, 1994), pp. 124–134.
[9] C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. **59**, 2044 (1987).
[10] Z. Y. Ou and L. Mandel, Phys. Rev. Lett. **61**, 50 (1988).
[11] X. Y. Zou, L. J. Wang, and L. Mandel, Phys. Rev. Lett. **67**, 318 (1991).
[12] L. J. Wang, X. Y. Zou, and L. Mandel, Phys. Rev. A **44**, 4614 (1991).
[13] R. A. Campos, B. E. A. Saleh, and M. C. Teich, Phys. Rev. A **40**, 1371 (1989).
[14] B. Yurke and M. Potasek, Phys. Rev. A **36**, 3464 (1987).
[15] R. H. Brown and R. Q. Twiss, Nature (London) **177**, 27 (1956).
[16] L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics* (Cambridge University Press, New York, 1995).
[17] M. Zukowski, A. Zeilinger, and H. Weinfurter, in *Fundamental Problems in Quantum Theory,* edited by D. M. Greenberg and A. Zeilinger [Ann. N.Y. Acad. Sci. **755**, 91 (1995)].
[18] J. G. Rarity, Ann. N.Y. Acad. Sci. **755**, 624 (1995).
[19] Z. Y. Ou, Quantum Semiclass. Opt. **9**, 599 (1997).
[20] Z. Y. Ou, J.-K. Rhee, and L. J. Wang, Phys. Rev. A **60**, 593 (1999).