

### Experiment on nonclassical fourth-order interference

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A new fourth-order interference experiment has been carried out and analyzed theoretically in classical and in quantum terms. Two photons produced in the process of parametric down-conversion provide the two inputs to a Mach-Zehnder type of interferometer, while two photodetectors coupled to a coincidence counter measure the output. The coincidence rate, after subtraction of accidentals, exhibits a cosine variation with the optical path difference, in agreement with quantum mechanics, but in disagreement with a classical analysis. By contrast, when two coherent light beams from a He:Ne laser are used as inputs to the interferometer, no fourth-order interference is observed.

#### I. INTRODUCTION

A number of optical interference experiments have recently been performed in which the field is in a nonclassical state.<sup>1-5</sup> The resulting interference patterns exhibit certain explicitly quantum-mechanical features. For example, in some fourth-order interference experiments with photon pairs the observed visibility of the interference was substantially greater than 50%, whereas classical optics allows it to be no larger than 50%. Perhaps even more striking were the results of two recent experiments<sup>4,5</sup> with down-converted photon pairs, for which classical optics predicts virtually no interference under the given experimental conditions.

In the following we report on another fourth-order interference experiment with nonclassical light, for which classical wave optics predicts no interference at all. The experiment is based on photon coincidence detection at the two outputs of a Mach-Zehnder type of interferometer. Two photons produced simultaneously in the process of parametric down-conversion, or frequency splitting, of light provide the two inputs to the interferometer, and the photons emerging simultaneously at the two outputs are registered by two detectors. The rate of simultaneous detection by both detectors in coincidence is found to exhibit a cosine dependence on the optical path difference, despite the fact that the two inputs are mutually incoherent and the two average output intensities do not vary with path difference. The experiment therefore violates the laws of classical optics. Finally, we compare the results of experiments performed with classical light wave inputs to the interferometer and show that the corresponding coincidence counting rate exhibits no fourth-order interference.

#### II. CLASSICAL TREATMENT OF THE EXPERIMENT

We consider the experimental arrangement shown in Fig. 1, in which two beam splitters BSI and BSO are used in combination to form a Mach-Zehnder type of interferometer. Let  $V_0(t), V_1(t)$  be complex analytic signals representing the two stationary input waves. Waves

represented by  $V_2(t)$  and  $V_3(t)$  emerge from BSI, and after time delays  $\tau_2$  and  $\tau_3$ , which may differ slightly, are introduced in the two interferometer arms, the two waves are combined at the output beam splitter BSO. The two output waves  $V_4(t)$  and  $V_5(t)$  then fall on two photodetectors D4 and D5, respectively, whose output pulses are fed to a coincidence counter that registers simultaneous detections. For simplicity we take the two beam splitters to be identical with 50% transmissivity and 50% reflectivity.

Because of the 90° phase shift of the reflected wave relative to the transmitted wave introduced by a symmetric beam splitter,<sup>6</sup> we may relate the various light waves as follows:

$$V_2(t) = \frac{1}{\sqrt{2}} [V_0(t) + iV_1(t)] , \tag{1}$$

$$V_3(t) = \frac{1}{\sqrt{2}} [iV_0(t) + V_1(t)] ,$$

$$V_4(t) = \frac{1}{\sqrt{2}} [V_2(t - \tau_2) + iV_3(t - \tau_3)] , \tag{2}$$

$$V_5(t) = \frac{1}{\sqrt{2}} [iV_2(t - \tau_2) + V_3(t - \tau_3)] .$$

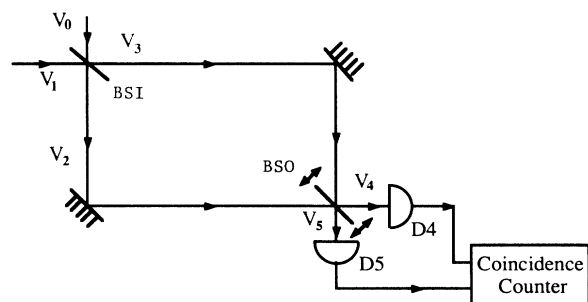


FIG. 1. The principle of the experiment under discussion.

By combining Eqs. (1) and (2), we obtain

$$\begin{aligned} V_4(t) &= \frac{1}{2}[V_0(t-\tau_2) - V_0(t-\tau_3) \\ &\quad + iV_1(t-\tau_2) + iV_1(t-\tau_3)], \\ V_5(t) &= \frac{1}{2}[iV_0(t-\tau_2) + iV_0(t-\tau_3) \\ &\quad - V_1(t-\tau_2) + V_1(t-\tau_3)]. \end{aligned} \quad (3)$$

Now suppose that very small variations of  $\tau_2, \tau_3$  are introduced. We replace  $\tau_2, \tau_3$  by  $\tau_2 + x_2, \tau_3 + x_3$ , respectively, with the understanding that both  $|x_2|$  and  $|x_3|$  are very

small compared with the second-order coherence time  $T_c$ . Then we may put

$$\begin{aligned} V_0(t-\tau_2-x_2) &= V_0(t-\tau_2)e^{i\omega_0 x_2}, \\ V_1(t-\tau_3-x_3) &= V_1(t-\tau_3)e^{i\omega_1 x_3}, \end{aligned} \quad (4)$$

where  $\omega_0$  and  $\omega_1$  are the midfrequencies of  $V_0(t)$  and  $V_1(t)$ , which are taken to be distinct for the moment. From Eqs. (3) and (4) we obtain for the instantaneous light intensities,

$$\begin{aligned} I_4(t) = |V_4(t)|^2 &= \frac{1}{4}[I_0(t-\tau_2) + I_0(t-\tau_3) + I_1(t-\tau_2) + I_1(t-\tau_3) - V_0^*(t-\tau_2)V_0(t-\tau_3)e^{i\omega_0(x_3-x_2)} + \text{c.c.} \\ &\quad + V_1^*(t-\tau_2)V_1(t-\tau_3)e^{i\omega_1(x_3-x_2)} + \text{c.c.} + iV_0^*(t-\tau_2)V_1(t-\tau_2)e^{i(\omega_1-\omega_0)x_2} + \text{c.c.} \\ &\quad - iV_0^*(t-\tau_3)V_1(t-\tau_3)e^{i(\omega_1-\omega_0)x_3} + \text{c.c.} + iV_0^*(t-\tau_2)V_1(t-\tau_3)e^{i(\omega_1x_3-\omega_0x_2)} + \text{c.c.} \\ &\quad - iV_0^*(t-\tau_3)V_1(t-\tau_2)e^{i(\omega_1x_2-\omega_0x_3)} + \text{c.c.}] , \end{aligned} \quad (5)$$

$$\begin{aligned} I_5(t) = |V_5(t)|^2 &= \frac{1}{4}[I_0(t-\tau_2) + I_0(t-\tau_3) + I_1(t-\tau_2) + I_1(t-\tau_3) + V_0^*(t-\tau_2)V_0(t-\tau_3)e^{i\omega_0(x_3-x_2)} + \text{c.c.} \\ &\quad - V_1^*(t-\tau_2)V_1(t-\tau_3)e^{i\omega_1(x_3-x_2)} + \text{c.c.} + iV_0^*(t-\tau_2)V_1(t-\tau_2)e^{i(\omega_1-\omega_0)x_2} + \text{c.c.} \\ &\quad - iV_0^*(t-\tau_3)V_1(t-\tau_3)e^{i(\omega_1-\omega_0)x_3} + \text{c.c.} - iV_0^*(t-\tau_2)V_1(t-\tau_3)e^{i(\omega_1x_3-\omega_0x_2)} + \text{c.c.} \\ &\quad + iV_0^*(t-\tau_3)V_1(t-\tau_2)e^{i(\omega_1x_2-\omega_0x_3)} + \text{c.c.}] . \end{aligned} \quad (6)$$

The average counting rates  $R_4$  and  $R_5$  of detectors D4 and D5 are proportional to the expectations of  $\langle I_4(t) \rangle$  and  $\langle I_5(t) \rangle$ . Let us assume that the optical field is stationary, and introduce the definitions

$$\begin{aligned} \Gamma_{ij}^{(1,1)}(\tau) &\equiv \langle V_i^*(t)V_j(t+\tau) \rangle \quad (i, j=0, 1), \\ \Gamma_{ij}^{(2,0)}(\tau) &\equiv \langle V_i^*(t)V_j^*(t+\tau) \rangle \quad (i, j=0, 1). \end{aligned} \quad (7)$$

Then Eqs. (5) and (6) yield

$$\begin{aligned} \langle I_4 \rangle &= \frac{1}{2}\langle I_0 \rangle + \frac{1}{2}\langle I_1 \rangle - \frac{1}{4}\Gamma_{00}^{(1,1)}(\tau_2-\tau_3)e^{i\omega_0(x_3-x_2)} + \text{c.c.} + \frac{1}{4}\Gamma_{11}^{(1,1)}(\tau_2-\tau_3)e^{i\omega_1(x_3-x_2)} + \text{c.c.} \\ &\quad + \frac{i}{4}\Gamma_{01}^{(1,1)}(0)(e^{i(\omega_1-\omega_0)x_2} - e^{i(\omega_1-\omega_0)x_3}) + \text{c.c.} + \frac{i}{4}\Gamma_{01}^{(1,1)}(\tau_2-\tau_3)e^{i(\omega_1x_3-\omega_0x_2)} + \text{c.c.} \\ &\quad - \frac{i}{4}\Gamma_{01}^{(1,1)}(\tau_3-\tau_2)e^{i(\omega_1x_2-\omega_0x_3)} + \text{c.c.} \end{aligned} \quad (8)$$

When  $\omega_1 = \omega_0$ , the terms in  $\Gamma_{01}^{(1,1)}(0)$  drop out. A similar result is obtained for  $\langle I_5 \rangle$ , except that the signs of the  $\Gamma^{(1,1)}$  terms are reversed.

Now suppose that input beam 1 is blocked momentarily, so that  $V_1 = 0$ , and yet no interference is registered by detectors D4 or D5 when  $x_2, x_3$  are varied. Then it follows from Eq. (8) that

$$\Gamma_{00}^{(1,1)}(\tau_2-\tau_3) = 0, \quad (9)$$

which implies that  $|\tau_2 - \tau_3|$  exceeds the coherence time  $T_c$  of the light. Similarly, we conclude, when input beam 0 is momentarily blocked and no interference is registered by the detectors when  $x_2, x_3$  are varied, that

$$\Gamma_{11}^{(1,1)}(\tau_2-\tau_3) = 0. \quad (10)$$

If the input waves  $V_0(t)$  and  $V_1(t)$  are mutually incoherent to the second order, then

$$\Gamma_{01}^{(1,1)}(\tau) = 0 \quad (11)$$

for all arguments. It then follows from Eq. (8) that when both inputs are present no second-order interference effect is expected to be registered by detectors D4 and D5 as  $x_2, x_3$  are varied.

Next we calculate the two-time intensity cross-correlation function

$$\Gamma_{45}^{(2,2)}(\tau) = \langle I_4(t)I_5(t+\tau) \rangle \quad (12)$$

between the two interferometer outputs. This is proportional to the joint probability that a detection is registered by D4 at time  $t$  and another detection by D5 at

time  $t + \tau$ . The average counting rate  $\mathcal{R}_{45}$  of the coincidence counter is given by the integral of  $\Gamma_{45}^{(2,2)}(\tau)$  with respect to  $\tau$  over the resolving time  $T_R$  of the coincidence counter,

$$\mathcal{R}_{45} = \int_{-T_R/2}^{T_R/2} d\tau \Gamma_{45}^{(2,2)}(\tau). \quad (13)$$

Because of the lengths of the expressions for  $I_4(t)$  and  $I_5(t)$  given by Eqs. (5) and (6), we shall not write out the

full expression for  $\Gamma_{45}^{(2,2)}(\tau)$ . Our interest centers mainly on the interference terms involving factors like

$$\exp[i(\omega_0 + \omega_1)(x_3 - x_2)],$$

after the integration in Eq. (13) is carried out.

Let us first suppose that input  $V_1$  is blocked momentarily. Then the fourth-order interference terms are of the form

$$\begin{aligned} & \{ \langle [I_0(t - \tau_2) + I_0(t - \tau_3)] V_0^*(t - \tau_2 + \tau) V_0(t - \tau_3 + \tau) \rangle \\ & - \langle [I_0(t - \tau_2 + \tau) + I_0(t - \tau_3 + \tau)] V_0^*(t - \tau_2) V_0(t - \tau_3) \rangle \} e^{i\omega_0(x_3 - x_2)} + \text{c.c.} \\ & - \langle V_0^*(t - \tau_2) V_0(t - \tau_3) V_0^*(t - \tau_2 + \tau) V_0(t - \tau_3 + \tau) \rangle e^{2i\omega_0(x_3 - x_2)} + \text{c.c.}, \end{aligned}$$

and similar expressions are encountered when input  $V_0$  is blocked instead. Because  $|\tau_2 - \tau_3|$  greatly exceeds the coherence time  $T_c$ , there will be no phase correlation between  $V_0(t - \tau_2)$  and  $V_0(t - \tau_3)$  and no phase correlation between  $V_0^*(t - \tau_2) V_0^*(t - \tau_2 + \tau)$  and  $V_0(t - \tau_3) V_0(t - \tau_3 + \tau)$ . Hence all the fourth-order interference terms vanish when one or the other input is blocked.

Next let us concentrate on the fourth-order cross correlations between  $V_0(t)$  and  $V_1(t)$  that contribute to interference in  $\Gamma_{45}^{(2,2)}(\tau)$  when both inputs are nonzero. As  $V_0(t)$  and  $V_1(t)$  are mutually incoherent, we expect any average in which either  $V_0(t)$  or  $V_1(t)$  is unpaired to vanish. The remaining interference terms in  $\Gamma_{45}^{(2,2)}(\tau)$  are of the form

$$\begin{aligned} \mathcal{J} = & \{ \langle V_0^*(t - \tau_2) V_0(t - \tau_3) V_1^*(t - \tau_2 + \tau) V_1(t - \tau_3 + \tau) \rangle + \langle V_1^*(t - \tau_2) V_1(t - \tau_3) V_0^*(t - \tau_2 + \tau) V_0(t - \tau_3 + \tau) \rangle \\ & + \langle V_0^*(t - \tau_2) V_1(t - \tau_3) V_0(t - \tau_3 + \tau) V_1^*(t - \tau_2 + \tau) \rangle \} e^{i(\omega_0 + \omega_1)(x_3 - x_2)} + \text{c.c.} \\ & + \langle V_0^*(t - \tau_2) V_1(t - \tau_3) V_0^*(t - \tau_2 + \tau) V_1(t - \tau_3 + \tau) \rangle e^{2i(\omega_1 x_3 - \omega_0 x_2)} + \text{c.c.} \\ & - \langle V_0(t - \tau_3) V_1^*(t - \tau_2) V_0(t - \tau_3 + \tau) V_1^*(t - \tau_2 + \tau) \rangle e^{2i(\omega_0 x_3 - \omega_1 x_2)} + \text{c.c.} \end{aligned} \quad (14)$$

Because of the lack of phase correlation between  $V_0(t)$  and  $V_1(t)$ , it might be expected that all the fourth-order correlations in Eq. (14) factor into the product of two second-order correlations, in the form

$$\begin{aligned} \mathcal{J} = & [2\Gamma_{00}^{(1,1)}(\tau_2 - \tau_3)\Gamma_{11}^{(1,1)}(\tau_2 - \tau_3) + \Gamma_{00}^{(1,1)}(\tau_2 - \tau_3 + \tau)\Gamma_{11}^{(1,1)}(\tau_2 - \tau_3 + \tau)] e^{i(\omega_0 + \omega_1)(x_3 - x_2)} + \text{c.c.} \\ & + \Gamma_{00}^{(2,0)}(\tau)\Gamma_{11}^{(0,2)}(\tau) e^{2i(\omega_1 x_3 - \omega_0 x_2)} + \text{c.c.} - \Gamma_{00}^{(0,2)}(\tau)\Gamma_{11}^{(2,0)}(\tau) e^{2i(\omega_0 x_3 - \omega_1 x_2)} + \text{c.c.} \end{aligned} \quad (15)$$

As  $\tau_2 - \tau_3$  exceeds  $T_c$ , each  $\Gamma^{(1,1)}$  term is zero and the remaining terms vanish because  $\Gamma_{00}^{(2,0)}(\tau) = 0 = \Gamma_{11}^{(2,0)}(\tau)$  for a stationary field.

Actually, because of the possibility that the product of two functions of  $t$  may have a correlation time that is substantially longer than that of either function separately, this argument leading to Eq. (15) is not always valid. The problem has been analyzed more carefully in a recent paper<sup>7</sup> dealing with another experiment, where it is shown that the contribution to  $\mathcal{R}_{45}$  of interference by terms of the form encountered in Eq. (14) is negligibly small when  $T_c \ll T_R$ . It then follows that under these conditions the coincidence rate  $\mathcal{R}_{45}$  is expected to exhibit no interference as the optical path indifference is changed.

Next let us consider a different situation. Suppose that the two input waves  $V_0(t)$  and  $V_1(t)$  are completely correlated, with

$$V_1(t) = V_0(t) e^{i\phi},$$

where  $\phi$  is some constant phase and the coherence time

$T_c$  is very long compared with all delays  $\tau_2, \tau_3$ . Moreover, let the intensity

$$I_0(t) = |V_0(t)|^2 = I_1(t)$$

be free from fluctuations. This might be the situation if  $V_0(t)$  and  $V_1(t)$  were derived from splitting a laser beam into two parts. Then it follows when we examine Eqs. (5) and (6) for  $I_4(t)$  and  $I_5(t)$  that all randomness disappears from the equations, and  $I_4(t)$  and  $I_5(t)$  become indistinguishable from  $\langle I_4 \rangle$  and  $\langle I_5 \rangle$ . Both of them exhibit a sinusoidal variation with optical path difference  $c(x_3 - x_2)$ . For the same reason also

$$\Gamma_{45}^{(2,2)}(\tau) = \langle I_4(t) I_5(t + \tau) \rangle$$

is indistinguishable from  $\langle I_4 \rangle \langle I_5 \rangle$ , and

$$\mathcal{R}_{45} = \int_{-T_R/2}^{T_R/2} \Gamma_{45}^{(2,2)}(\tau) d\tau = \langle I_4 \rangle \langle I_5 \rangle T_R. \quad (16)$$

But the left side of this equation is a measure of the average counting rate of the coincidence detector, whereas the right side is a measure of the accidental coincidence

rate  $\mathcal{R}_A$ , due to the accidental arrival of uncorrelated pulses from D4 and D5 within the resolving time  $T_R$ . It then follows that  $\mathcal{R}_{45} - \mathcal{R}_A = 0$ , and any interference effects that show up in  $\mathcal{R}_{45}$  disappear when we subtract accidental coincidences. Hence there is again no fourth-order interference effect. The situation is quite different for a quantum field, as we now show.

### III. QUANTUM TREATMENT WITH TWO-PHOTON INPUT

For the quantum treatment we consider the inputs to the interferometer to consist of one photon at port 0 and one photon at port 1, which are produced simultaneously in the process of spontaneous parametric down-conversion in a nonlinear medium. This process was the basis for a number of recent fourth-order, nonclassical interference experiments,<sup>2-5</sup> and it has been treated theoretically several times recently.<sup>8-10</sup>

In order to illustrate the main difference between the experiment with two photons and with two classical waves, let us first adopt a grossly oversimplified point of view, in which all fields are monochromatic. Corresponding to the classical Eqs. (2), we have the operator equations for modes 2,3,4,5,

$$\begin{aligned}\hat{a}_4 &= \frac{1}{\sqrt{2}}(\hat{a}_2 e^{i\phi_2} + i\hat{a}_3 e^{i\phi_3}), \\ \hat{a}_5 &= \frac{1}{\sqrt{2}}(i\hat{a}_2 e^{i\phi_2} + \hat{a}_3 e^{i\phi_3}),\end{aligned}\quad (17)$$

in which  $\phi_2, \phi_3$  are the phase shifts introduced in the two interferometer arms. Now it has been shown that when two similar photons enter the input beam splitter at ports 0 and 1, two photons always emerge together either at port 2 or at port 3,<sup>11,12</sup> so that modes 2 and 3 are in the superposition state

$$|\psi\rangle = \frac{1}{\sqrt{2}}(|2\rangle_2|0\rangle_3 + |0\rangle_2|2\rangle_3).$$

For this state we readily obtain, with the help of Eqs. (17), for the expected number of detections by detector D4 or D5,

$$\langle \psi | \hat{a}_4^\dagger \hat{a}_4 | \psi \rangle = 1 = \langle \psi | \hat{a}_5^\dagger \hat{a}_5 | \psi \rangle, \quad (18a)$$

and for the joint detections by D4 and D5,

$$\langle \psi | \hat{a}_4^\dagger \hat{a}_5^\dagger \hat{a}_5 \hat{a}_4 | \psi \rangle = \frac{1}{2}[1 + \cos 2(\phi_2 - \phi_1)]. \quad (18b)$$

Hence there is no second-order interference, but there is fourth-order interference with 100% visibility. This is quite different from the classical situation treated in Sec. II, and it is a consequence of the interference of photon pairs, rather than single photons, in the interferometer.

However, the down-converted photons are far from monochromatic. The two photons are also entangled with each other and with the vacuum,<sup>8,9</sup> so that our quantum calculation needs to be repeated under more realistic assumptions. If we suppose that all signal ( $s$ ) and idler ( $i$ ) down-converted photons are collected by the apparatus, so that directional effects can be ignored, and the initial signal-idler field is in the vacuum state at time

$t=0$ , then the state at a later time  $t$ , which is short compared with the average time interval between down-conversions, can be well approximated by<sup>9</sup>

$$\begin{aligned}|\psi(t)\rangle &= \left[ 1 + \eta V \delta\omega \sum_{\omega'} \sum_{\omega''} \phi(\omega', \omega'') \frac{\sin \frac{1}{2}(\omega' + \omega'' - \omega_p)t}{\frac{1}{2}(\omega' + \omega'' - \omega_p)} \right. \\ &\quad \times e^{i(\omega' + \omega'' - \omega_p)t/2} \\ &\quad \left. \times \hat{a}_s^\dagger(\omega') \hat{a}_i^\dagger(\omega'') + \text{H.c.} \right] \\ &\quad \times |\text{vac}\rangle_s |\text{vac}\rangle_i.\end{aligned}\quad (19)$$

We have used a discrete-mode decomposition with mode spacing  $\delta\omega$ , and in the limit  $\delta\omega \rightarrow 0$  sums over  $\omega$  become integrals.  $\phi(\omega', \omega'')$  is a symmetric weight function that depends on the nonlinear  $\chi^{(2)}$  susceptibility and characterizes the spectrum of the down-converted light. It is normalized so that

$$2\pi\delta\omega \sum_{\omega} |\phi(\omega, \omega_p - \omega)|^2 = 1. \quad (20)$$

$\omega_p$  is the frequency of the pump light and  $V$  is its complex amplitude.  $\eta$  is a parameter that represents the efficiency of down-conversion, such that if  $|V|^2$  is the rate at which pump photons are incident on the nonlinear medium, then  $|\eta V|^2$  is the rate at which down-converted photons are produced.

In the following we regard the state  $|\psi(t)\rangle$  given by Eq. (19) as the input state to the Mach-Zehnder interferometer, with the signal photon corresponding to port 0 in Fig. 1 and the idler photon corresponding to port 1. The fields  $\hat{E}_2^{(+)}(t)$  and  $\hat{E}_3^{(+)}(t)$  on the output side of the 50%:50% input beam splitter BS1 can then be given the mode expansions<sup>9</sup>

$$\begin{aligned}\hat{E}_2^{(+)}(t) &= \left[ \frac{\delta\omega}{2\pi} \right]^{1/2} \frac{1}{\sqrt{2}} \sum_{\omega_2} [\hat{a}_0(\omega_2) + i\hat{a}_1(\omega_2)] e^{-i\omega_2 t}, \\ \hat{E}_3^{(+)}(t) &= \left[ \frac{\delta\omega}{2\pi} \right]^{1/2} \frac{1}{\sqrt{2}} \sum_{\omega_3} [i\hat{a}_0(\omega_3) + \hat{a}_1(\omega_3)] e^{-i\omega_3 t}.\end{aligned}\quad (21)$$

These quantum relations correspond to the classical Eqs. (1). After suitable propagation delays  $\tau_2$  and  $\tau_3$ , the fields are again mixed at the output beam splitter BSO shown in Fig. 1, such that the fields emerging from the interferometer are represented by

$$\begin{aligned}\hat{E}_4^{(+)}(t) &= \left[ \frac{\delta\omega}{2\pi} \right]^{1/2} \frac{1}{\sqrt{2}} \\ &\quad \times \sum_{\omega_4} [\hat{a}_2(\omega_4) e^{i\omega_4\tau_2} + i\hat{a}_3(\omega_4) e^{i\omega_4\tau_3}] e^{-i\omega_4 t}, \\ \hat{E}_5^{(+)}(t) &= \left[ \frac{\delta\omega}{2\pi} \right]^{1/2} \frac{1}{\sqrt{2}} \\ &\quad \times \sum_{\omega_5} [i\hat{a}_2(\omega_5) e^{i\omega_5\tau_2} + \hat{a}_3(\omega_5) e^{i\omega_5\tau_3}] e^{-i\omega_5 t}.\end{aligned}\quad (22)$$

Here  $\hat{a}_2(\omega)$ ,  $\hat{a}_3(\omega)$  are the mode amplitudes of fields  $\hat{E}_2^{(+)}(t)$ ,  $\hat{E}_3^{(+)}(t)$  given by Eqs. (21). On combining Eqs. (21) and (22), we arrive at

$$\begin{aligned}\hat{E}_4^{(+)}(t) &= \left(\frac{\delta\omega}{2\pi}\right)^{1/2} \frac{1}{2} \sum_{\omega_4} [\hat{a}_0(\omega_4)(e^{i\omega_4\tau_2} - e^{i\omega_4\tau_3}) + i\hat{a}_1(\omega_4)(e^{i\omega_4\tau_2} + e^{i\omega_4\tau_3})] e^{-i\omega_4 t}, \\ \hat{E}_5^{(+)}(t) &= \left(\frac{\delta\omega}{2\pi}\right)^{1/2} \frac{1}{2} \sum_{\omega_5} [i\hat{a}_0(\omega_5)(e^{i\omega_5\tau_2} + e^{i\omega_5\tau_3}) + \hat{a}_1(\omega_5)(-e^{i\omega_5\tau_2} + e^{i\omega_5\tau_3})] e^{-i\omega_5 t}\end{aligned}\quad (23)$$

We may now use Eqs. (17) and (21) to calculate the rate of photodetection by detector D4 at time  $t$ , which is proportional to

$$\begin{aligned}R_4(t) &= \langle \psi(t) | \hat{E}_4^{(-)}(t) \hat{E}_4^{(+)}(t) | \psi(t) \rangle \\ &= \left(\frac{\delta\omega}{2\pi}\right)^2 \frac{1}{4} |\eta V|^2 (\delta\omega)^2 \sum_{\omega'} \sum_{\omega''} \sum_{\omega'''} \sum_{\omega''''} \sum_{\omega_4} \sum_{\omega'_4} \phi^*(\omega', \omega'') \phi(\omega''', \omega'''' ) \\ &\quad \times \frac{\sin(\omega' + \omega'' - \omega_p)t/2}{(\omega' + \omega'' - \omega_p)/2} e^{-i(\omega' + \omega'' - \omega_p)t/2} \\ &\quad \times \frac{\sin(\omega''' + \omega'''' - \omega_p)t/2}{(\omega''' + \omega'''' - \omega_p)/2} e^{i(\omega''' + \omega'''' - \omega_p)t/2} e^{i(\omega_4 - \omega'_4)t} \\ &\quad \times \langle \omega' | \langle \omega'' | [\hat{a}_0^\dagger(\omega_4)(e^{-i\omega_4\tau_2} - e^{-i\omega_4\tau_3}) - i\hat{a}_1^\dagger(\omega_4)(e^{-i\omega_4\tau_2} + e^{-i\omega_4\tau_3})] \\ &\quad \times [\hat{a}_0(\omega'_4)(e^{i\omega'_4\tau_2} - e^{i\omega'_4\tau_3}) + i\hat{a}_1(\omega'_4)(e^{i\omega'_4\tau_2} + e^{i\omega'_4\tau_3})] | \omega'''' \rangle_0 | \omega'''' \rangle_1. \quad (24)\end{aligned}$$

The matrix element  $M$  yields

$$\begin{aligned}M &= (e^{i\omega'''\tau_2} - e^{i\omega'''\tau_3}) \\ &\quad \times (e^{-i\omega'\tau_2} - e^{-i\omega'\tau_3}) \delta_{\omega'\omega_4} \delta_{\omega''\omega'_4} \delta_{\omega'''\omega_4} \delta_{\omega''''\omega'_4} \\ &\quad + (e^{i\omega'''\tau_2} + e^{i\omega'''\tau_3}) \\ &\quad \times (e^{-i\omega'\tau_2} + e^{-i\omega'\tau_3}) \delta_{\omega''\omega_4} \delta_{\omega'''\omega'_4} \delta_{\omega''''\omega'_4}.\end{aligned}$$

We now substitute this result into Eq. (24) and introduce changes of variables of the form

$$\omega' = \omega_p - \omega'' + \Omega',$$

etc. We let  $\delta\omega \rightarrow 0$  so that the sum over  $\Omega'$  converts to an integral, and we assume that  $\phi^*(\omega', \omega'')$  is a sufficiently slowly varying function of  $\omega'$ , so that for large  $t$  we may write

$$\phi^*(\omega_p - \omega'' + \Omega', \omega'') \approx \phi^*(\omega_p - \omega'', \omega'').$$

We also note that

$$\begin{aligned}\frac{1}{2\pi} \int_{\omega'' - \omega_p}^{\infty} d\Omega' \frac{\sin(\Omega't/2)}{\Omega'/2} e^{\pm i\Omega'(t/2 - \tau)} \\ \approx \frac{1}{2\pi} \int_{-\infty}^{\infty} d\Omega' \frac{\sin(\Omega't/2)}{\Omega'/2} e^{\pm i\Omega'(t/2 - \tau)} = \Theta(\tau; t),\end{aligned}\quad (25)$$

where  $\Theta(\tau; t)$  is the unit step function defined by

$$\Theta(\tau; t) \begin{cases} \cong 1 & \text{for } 0 < \tau < t \\ \cong 0 & \text{for } \tau < 0, \tau > t. \end{cases} \quad (26)$$

We then find that provided  $t > \tau_2, \tau_3$  all dependence on  $\tau_2, \tau_3$  disappears from Eq. (24), and we are left with

$$\begin{aligned}R_4(t) &= 2\pi \frac{\delta\omega}{4} |\eta V|^2 \sum_{\omega'} \phi^*(\omega_p - \omega'', \omega'') \phi(\omega_p - \omega'', \omega'') \times 4 \\ &= |\eta V|^2,\end{aligned}\quad (27)$$

when we make use of Eq. (20). Similarly we may show that the photodetection rate at D5 is proportional to

$$R_5(t) = |\eta V|^2. \quad (28)$$

It follows that no second-order interference effect shows up in the experiment illustrated in Fig. 1 when two down-converted photons serve as inputs to the interferometer. If we compare this conclusion with the classical result given by Eq. (8), we see that it corresponds to the classical conditions

$$\begin{aligned}\Gamma_{00}^{(1,1)}(\tau_2 - \tau_3) = 0 = \Gamma_{11}^{(1,1)}(\tau_2 - \tau_3), \\ \Gamma_{01}^{(1,1)}(\tau) = 0.\end{aligned}\quad (29)$$

The first condition is a consequence of the very short coherence time  $T_c$ , and the second condition, implying absence of mutual coherence, is connected with the fact that single photons have no definite phase.

Next we turn to the calculation of the joint probability of detecting a photon with detector D4 at time  $t$  and with detector D5 at time  $t + \tau$ . Provided that the probability of two down-conversions is much smaller than that for one, the required probability is proportional to<sup>9</sup>

$$\mathcal{P}_{45}(t, t + \tau) = \langle \psi(t) | \hat{E}_4^{(-)}(t) \hat{E}_5^{(-)}(t + \tau) \hat{E}_5^{(+)}(t + \tau) \hat{E}_4^{(+)}(t) | \psi(t) \rangle. \quad (30)$$

After substituting from Eq. (19) for  $|\psi(t)\rangle$  and from Eqs. (23) for  $\hat{E}_4^{(+)}(t)$  and  $\hat{E}_5^{(+)}(t)$ , and evaluating the matrix element, we arrive at

$$\begin{aligned} \mathcal{P}_{45}(t, t + \tau) &= \frac{\delta\omega^4}{(2\pi)^2} \frac{|\eta V|^2}{16} \sum_{\omega'} \sum_{\omega''} \sum_{\omega'''} \sum_{\omega''''} \phi^*(\omega', \omega'') \phi(\omega''', \omega'''' ) \\ &\quad \times \frac{\sin(\omega' + \omega'' - \omega_p)t/2}{(\omega' + \omega'' - \omega_p)/2} \frac{\sin(\omega''' + \omega'''' - \omega_p)t/2}{(\omega''' + \omega'''' - \omega_p)/2} \\ &\quad \times e^{i(\omega' + \omega'' - \omega''' - \omega'''' )t/2} (e^{i(\omega'' - \omega'''' )\tau} + e^{i(\omega' - \omega'''' )\tau} + e^{i(\omega'' - \omega''')\tau} + e^{i(\omega' - \omega''')\tau}) \\ &\quad \times [F_-^*(\omega') F_-^*(\omega'') + F_+^*(\omega') F_+^*(\omega'')] [F_+(\omega''') F_+(\omega'''' ) + F_-(\omega''') F_-(\omega'''' )], \end{aligned} \quad (31)$$

where we have written

$$F_{\pm}(\omega) \equiv e^{i\omega\tau_2} \pm e^{i\omega\tau_3}. \quad (32)$$

We again introduce changes of variables  $\omega' + \omega'' - \omega_p = \Omega''$ , etc., as above, and make use of the integral relation (25). Then provided  $\tau_2$  and  $\tau_3$  are long compared with the coherence time  $T_c$  of the incident light, we may drop all the  $\Theta(\tau_2 - \tau; t)$ ,  $\Theta(\tau_3 - \tau; t)$  functions, and obtain

$$\begin{aligned} \mathcal{P}_{45}(t, t + \tau) &= \frac{(2\pi)^2 |\eta V|^2}{4} |\gamma(\tau)|^2 |e^{i\omega_p\tau_2} + e^{i\omega_p\tau_3}|^2 \\ &= \frac{(2\pi)^2 |\eta V|^2}{2} |\gamma(\tau)|^2 [1 + \cos\omega_p(\tau_2 - \tau_3)]. \end{aligned} \quad (33)$$

We have written

$$\gamma(\tau) \equiv \frac{1}{2\pi} \int_0^\infty \phi(\omega, \omega_p - \omega) e^{i\omega\tau} d\omega \quad (34)$$

for the autocorrelation function associated with the spectral function  $\phi(\omega, \omega_p - \omega)$  of the down-converted photons. Because of the wide bandwidth of the weight function  $\phi(\omega, \omega_p - \omega)$  which is symmetric and centered on  $\omega = \omega_p/2$ ,  $|\gamma(\tau)|$  has a very short range  $T_c$ , typically less than a picosecond.

As before, the average coincidence rate measured by the coincidence counter is proportional to the integral of  $\mathcal{P}_{45}(t, t + \tau)$  with respect to  $\tau$ , over the range  $-T_R/2$  to  $T_R/2$ . When  $T_R \gg T_c$ , this is equivalent to integrating  $\mathcal{P}_{45}(t, t + \tau)$  over the infinite range. With the help of the Plancherel theorem

$$\int_{-\infty}^{\infty} |\gamma(\tau)|^2 d\tau = \frac{1}{2\pi} \int_0^\infty |\phi(\omega, \omega_p - \omega)|^2 d\omega$$

and the relation (20), we then arrive at

$$\begin{aligned} \mathcal{R}_{45} &= \int_{-\infty}^{\infty} \mathcal{P}_{45}(t, t + \tau) d\tau \\ &= \frac{1}{2} |\eta V|^2 [1 + \cos\omega_p(\tau_2 - \tau_3)]. \end{aligned} \quad (35)$$

By making use of Eqs. (27) and (28), we can derive the accidental rate of coincidence counting, which is given by

$$\mathcal{R}_A = |\eta V|^4 T_R, \quad (36)$$

and we note that so long as  $|\eta V|^2 T_R \ll 1$ , this is negli-

ble compared with  $\mathcal{R}_{45}$  given by Eq. (35). In other words, when  $T_R$  is much shorter than the average time interval between down-conversions, the excess coincidence rate is expected to exhibit interference with 100% visibility, in agreement with the earlier equation (18b). This may be contrasted with the classical situation described by Eq. (14) that predicts no fourth-order interference.

#### IV. EXPERIMENT

We have tested some of the foregoing theoretical predictions experimentally. An outline of the setup for the first experiments is shown in Fig. 2. The two photons entering at the input to the (slightly unbalanced) Mach-Zehnder interferometer are produced by down-conversion in a nonlinear crystal of  $\text{LiIO}_3$ , which is pumped by an incident light beam at a wavelength of 351.1 nm from an argon ion laser. Down-converted and simultaneous signal and idler photons of about 700 nm wavelength emerge at relative angles of  $\pm 7^\circ$  from the crystal and provide the inputs to the input beam splitter BSI. This part of the apparatus is very similar to that used previously.<sup>11</sup> BSI is mounted on a micrometer that allows its position to be translated. It has been shown that when BSI is in the symmetric position, the two photons emerging from the output sides of BSI almost always appear together on one or the other side, and almost never on both sides simultaneously, because of destructive interference.<sup>11,12</sup> This principle is used to adjust BSI until it is symmetrically located with respect to the signal and idler photons. The light emerging from BSI passes through the interferometer and is eventually mixed by the output beam splitter BSO, whose output beams are directed to the two photon counting detectors D4 and D5. The path difference  $c(\tau_3 - \tau_2)$  through the interferometer, which is slightly unbalanced permanently, can be varied over a range of a few wavelengths by mounting BSO on a piezoelectric transducer and varying the applied voltage. A pinhole and an interference filter of bandwidth  $10^{12}$  Hz placed in front of each detector determine the angular spread and the frequency spread of the detected light. After amplification and pulse shaping, the photomultiplier pulses are fed to counters and to a computer-controlled coincidence counter that yields the coincidence counting rate. The resolving time  $T_R$  was made about 13 ns, but the precise value was determined

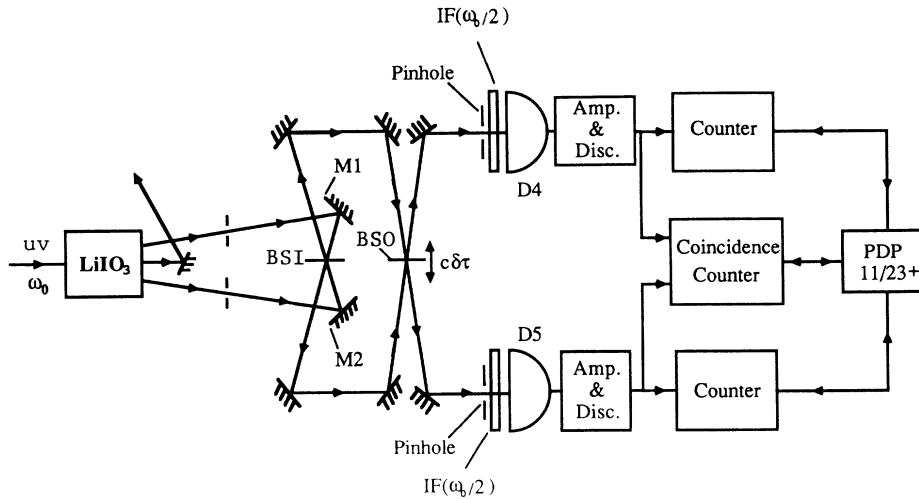


FIG. 2. Outline of the experimental setup.

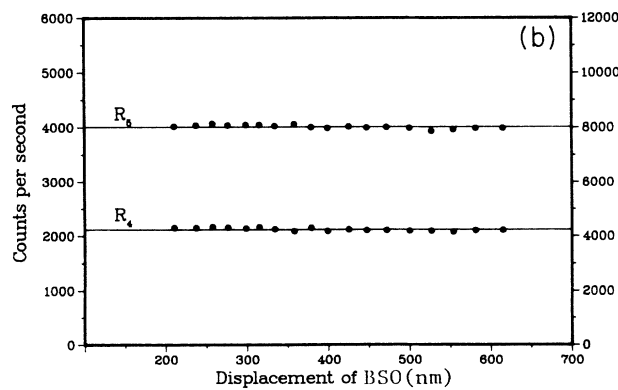
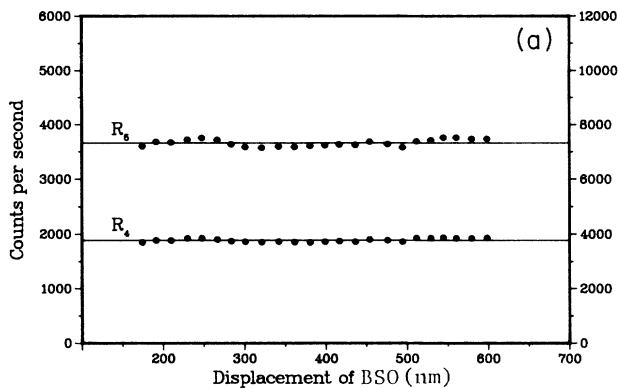


FIG. 3. The measured counting rates  $R_4, R_5$  of the two detectors as a function of optical path difference (a) when two photons enter simultaneously at ports 0 and 1 and (b) when the input to port 1 is blocked (and the pump power is increased somewhat). The standard deviations are smaller than the spot size. The scale on the left applies to  $R_5$  and that on the right to  $R_4$ .

by use of the relation  $\mathcal{R}_A = R_4 R_5 T_R$  for the accidental coincidence rate. Counts due to the detector dark currents were measured separately and subtracted out.

Figure 3(a) shows the measured counting rates  $R_4, R_5$  of detectors D4, D5 as a function of optical path difference  $c(\tau_3 - \tau_2)$ . Background counting rates of  $105 \text{ sec}^{-1}$  and  $115 \text{ sec}^{-1}$  have been subtracted out. It is evident that there is no second-order interference, as predicted by Eqs. (27) and (28). Also shown in Fig. 3(b) are the counting rates  $R_4, R_5$  when one or the other input to the input beam splitter BSI is blocked and the laser power is turned up a little. The absence of second-order

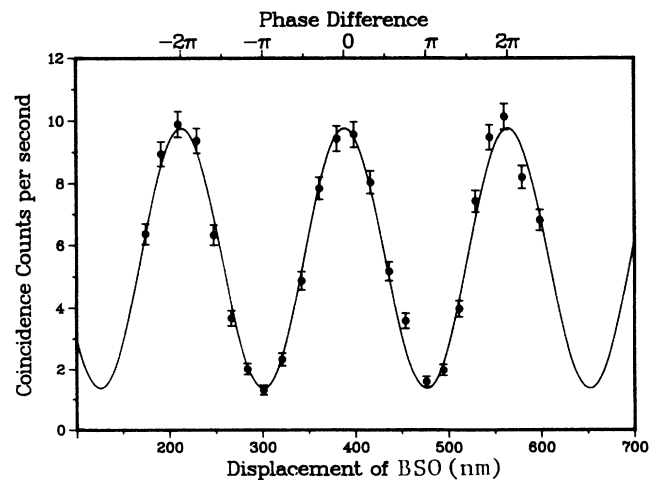


FIG. 4. The measured coincidence counting rate  $\mathcal{R}_{45}$ , after subtraction of accidentals, as a function of optical path difference with two simultaneous photons as inputs.

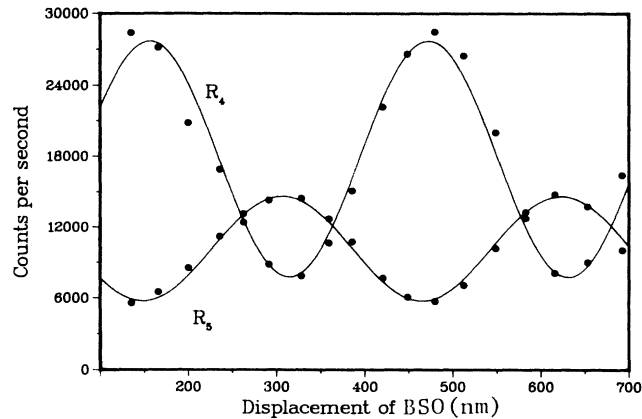


FIG. 5. The measured counting rates  $R_4, R_5$  of the two detectors as a function of optical path difference, with two mutually coherent laser beams as inputs. The standard deviations are smaller than the spot size.

interference in the last case is a reflection of the fact that the interferometer is unbalanced and the optical path difference  $c(\tau_2 - \tau_3)$  exceeds the coherence length  $cT_c$  when  $T_c \approx 10^{-12}$  sec. This experimental result is the basis of the conclusion leading to Eqs. (9) and (10) above. Figure 4 shows the measured coincidence rate  $\mathcal{R}_{45}$ , after subtraction of accidentals ( $\mathcal{R}_A \sim 0.17 \text{ sec}^{-1}$ ), as a function of the optical path difference  $c(\tau_3 - \tau_2)$  through the interferometer. This time we clearly have a fourth-order interference pattern, in agreement with the quantum prediction given by Eq. (35), although the visibility is a little less than 100%. The reason is probably connected with imperfections in the alignment. This result is, however, in violation of the conclusion resulting from the classical equation (14), which predicts no fourth-order interference for this case.

For comparison with the quantum effect just described, we then performed a second series of experiments, in which the inputs to the Mach-Zehnder interferometer at BSI were two classical light beams derived by splitting a He:Ne laser beam. The two input beams were therefore mutually coherent. Figure 5 shows the measured counting rates  $R_4, R_5$  of photodetectors D4, D5 as a function of the optical path difference (after subtraction of background rates of order  $150 \text{ sec}^{-1}$ ), and, as expected, second-order interference is observed. Figure 6 shows the measured coincidence rate, after subtraction of accidentals. Because  $R_4$  and  $R_5$  vary strongly with path difference, so does the accidental rate  $\mathcal{R}_A$ , with values oscillating between about  $1.1 \text{ sec}^{-1}$  and  $2.6 \text{ sec}^{-1}$ . However, once  $\mathcal{R}_A$  is subtracted out, we are left with near zero values of the coincidence rate. The standard deviations

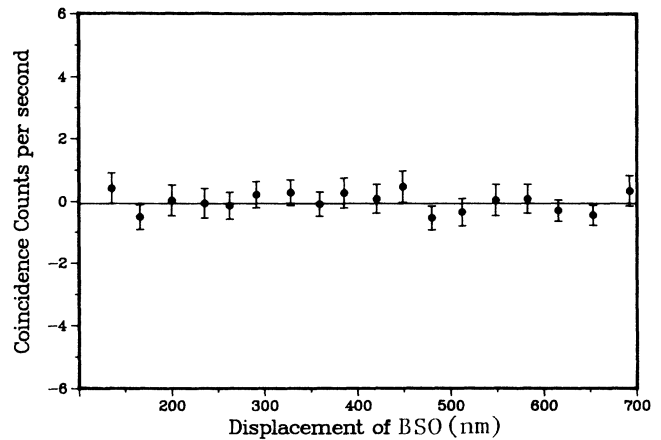


FIG. 6. The measured coincidence counting rate  $\mathcal{R}_{45}$ , after subtraction of accidentals  $\mathcal{R}_A$ , as a function of optical path difference, with two mutually coherent laser beams as inputs.

are relatively large, because it was found desirable to keep the measurement time per point to a few seconds in order to minimize the effect of slow phase drifts. As predicted by Eqs. (13) and (16), no fourth-order interference exists for this classical field. This further emphasizes that the effects shown in Fig. 4 are nonclassical and characteristic of a quantum field.

## V. DISCUSSION

We have demonstrated that under the conditions  $(\tau_2 - \tau_3) \gg T_c$ , when the entangled two-photon state produced in the down-conversion process is the input to the Mach-Zehnder interferometer, no second-order interference is observed, but there is fourth-order interference. On the other hand, when two mutually coherent beams from a He:Ne laser serve as inputs to the interferometer, then there is second-order but no fourth-order interference. To some extent the quantum state and the classical state behave as direct opposites.

One possible way to understand the difference between the two cases is to emphasize that with the down-converted light as input the interference is between photon pairs, whereas with two coherent or classical fields as input, we observe only one-photon interference. As in the Franson type of interference experiment,<sup>4,5</sup> it appears that no ergodic classical field can give rise to the interference effects observed with two down-converted photons.

## ACKNOWLEDGMENTS

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<sup>1</sup>P. Grangier, G. Roger, and A. Aspect, *Europhys. Lett.* **1**, 173 (1986).

<sup>2</sup>R. Ghosh and L. Mandel, *Phys. Rev. Lett.* **59**, 1903 (1987).

<sup>3</sup>Z. Y. Ou and L. Mandel, *Phys. Rev. Lett.* **62**, 2941 (1989).

<sup>4</sup>Z. Y. Ou, X. Y. Zou, L. J. Wang, and L. Mandel, *Phys. Rev. Lett.* **65**, 321 (1990).

<sup>5</sup>P. G. Kwiat, W. A. Vareka, C. K. Hong, H. Nathel, and R. Y. Chiao, *Phys. Rev. A* **41**, 2910 (1990).



<sup>6</sup>See, for example, M. Born and E. Wolf, *Principles of Optics*, 5th ed. (Pergamon, Oxford, 1975).

<sup>7</sup>Z. Y. Ou and L. Mandel, *J. Opt. Soc. Am. B* (to be published).

<sup>8</sup>P. Grangier, M. J. Potasek, and B. Yurke, *Phys. Rev. A* **38**, 3132 (1988).

<sup>9</sup>Z. Y. Ou, L. J. Wang, and L. Mandel, *Phys. Rev. A* **40**, 1428 (1989).

<sup>10</sup>Z. Y. Ou, L. J. Wang, X. Y. Zou, and L. Mandel, *Phys. Rev. A* **41**, 1597 (1990).

<sup>11</sup>C. K. Hong, Z. Y. Ou, and L. Mandel, *Phys. Rev. Lett.* **59**, 2044 (1987).

<sup>12</sup>Z. Y. Ou, C. K. Hong, and L. Mandel, *Opt. Commun.* **63**, 118 (1987).