First-order interference of nonclassical light emitted spontaneously at different times

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We study the first-order interference in spontaneous parametric down-conversion generated by two pump pulses that are well distinguishable in time. The observed modulation in the angular distribution of the signal photon can only be explained in terms of a quantum-mechanical description based on biphoton states. The condition for observing interference in the signal channel is shown to depend on the parameters of the idler photon.

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Nonclassical interference is one of the most remarkable phenomena in quantum optics. In particular, it can be observed in experiments with spontaneous parametric down-conversion (SPDC) [1], a nonlinear optical process in which high-energy pump photons are converted into pairs of low-energy photons (usually called signal and idler) inside a crystal with quadratic nonlinearity χ. It has been shown in many experiments that the quantum state of the signal-idler photon pair is entangled [2]. Due to the nonclassical correlation between the signal and the idler photons emitted in SPDC, the term ''biphoton'' has been suggested [3]. Many experiments have made use of SPDC to demonstrate fascinating topics in quantum optics, such as the test of Bell's inequalities, quantum communication, quantum teleportation, etc. [4], and its possible applications include quantum communication, computation, and cryptography [5]. All these experiments basically belong to the same category: quantum interference.

Among the variety of quantum interference experiments, there is a collection of work where the first-order interference is observed in the signal or in the idler of SPDC in which a pair may be emitted from two spatially separated domains [6–8]. This kind of interference is nonclassical: both the signal and the idler exhibit noise (thermal) statistics. From the classical point of view, spatially separated SPDC sources should exhibit no first-order interference. Indeed, one cannot observe stable first-order interference of light emitted by independent classical thermal sources, such as two similar light-emitting diodes [9]. Another interesting feature of the first-order interference of SPDC is that it depends on the phases of the pump, the signal, and the idler photons [10]. Moreover, the condition for the signal photon interference depends on the parameters of the idler photon [8].

In this Rapid Communication, we report an experimental observation of an alternative type of quantum interference, in which the sources of SPDC are separated not in space but in time, as SPDC is generated from two coherent femtosecond laser pulses [11,12]. Depending on the experimental conditions, interference is observed either in the angular distribution of the signal intensity (first-order interference) or in the coincidence counts from the detectors registering the signal and the idler photon (second-order interference). In this paper, we focus on the first-order interference; the second-order interference will be discussed elsewhere [13].

Let us consider type-II collinear degenerate SPDC generated in a BBO crystal of length L from two short pump pulses. The signal photon is separated from the idler and its intensity is measured by a detector that is placed after a narrow-band interference filter; see Fig.1. The pump field can be represented in the form

$$E_p(r,t)=\tilde{E}(t-z/u_p)\exp(-i\omega_p t+ik_p z), \tag{1}$$

where u_p is the pump group velocity inside the BBO, $\tilde{E}(t)$ is the envelope, and ω_p is the pump central frequency. We consider the pump pulse as quasimonochromatic since $\omega_p \gg \Delta\omega_p$, even for a femtosecond laser pulse. If the pump consists of two identical pulses separated in time by T_p, then $\tilde{E}(t)=E_0(t)+E_0(t+T_p)\exp(-i\omega_p T_p)$, where $E_0(t)$ is the single-pulse envelope.

In first-order perturbation theory, the quantum state of SPDC is given by [14]

$$|\Psi\rangle=|\mathrm{vac}\rangle+\sum_{k_s,k_i} F_{k_s,k_i}|1\rangle_{k_s}|1\rangle_{k_i}, \tag{2}$$

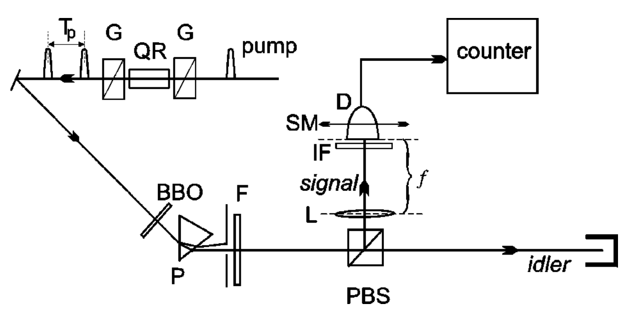

FIG. 1. Experimental setup. The incident single pump pulse is divided into two temporally separated pulses. Collinear degenerate type-II SPDC is generated from the BBO crystal pumped by the two pulses. Angular distribution of the signal photon is observed by scanning a detector in the focal plane of the lens. Interference filters (IF) are used for spectral selection of the signal.

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where the notation $|1\rangle_{k_s}|1\rangle_{k_i}$ means a two-photon state in the mode $\mathbf{k}_s, \mathbf{k}_i$, and the two-photon spectral function is given by

$$F_{k_s,k_i} = -\frac{i\chi}{\hbar}\int_{t_0}^{t} dt' \int_V d^3r \tilde{E}(t' - z/u_p) \times \exp\{-i(\omega_p - \omega_s - \omega_i)t' + i(\mathbf{k}_p - \mathbf{k}_s - \mathbf{k}_i)\cdot\mathbf{r}\}, \tag{3}$$

where the subscripts p, s, and i refer to the pump, the signal, and the idler, respectively. Since the pump pulse is bound in time, the integration over t' can be extended to infinite limits. As a result, it gives the Fourier transform of the pump envelope, $\tilde{E}(\omega_s + \omega_i - \omega_p)$, which in the cw case becomes a delta function, $\delta(\omega_s + \omega_i - \omega_p)$. We obtain

$$F_{k_s,k_i} = -\frac{2\pi i\chi}{\hbar}\tilde{E}(\omega_s + \omega_i - \omega_p)\delta(k_{sx} + k_{ix}) \times \mathrm{sinc}\left\{\frac{L}{2}\left(k_p - k_{sz} - k_{iz} + \frac{\omega_s + \omega_i - \omega_p}{u_p}\right)\right\}, \tag{4}$$

where, for example, k_{sx} and k_{sz} are the transverse and longitudinal components of the signal wave vector, respectively. For a two-pulse pump, the envelope spectrum is $\tilde{E}(\omega) = E_0(\omega)\cos\{(\omega - \omega_p)T_p/2\}$, with $E_0(\omega)$ denoting the Fourier transform of the single-pulse envelope.

The coincidence counting rate (probability of detecting a biphoton) is $R_c \propto |F_{k_s,k_i}|^2$. The single counting rate of the signal detector R_s is calculated by integrating R_c over all idler modes.

$$\begin{aligned} R_s &\propto \int dk_{iz} dk_{ix} |F_{k_s,k_i}|^2 \\ &= \frac{4\pi^2\chi^2 L^2}{\hbar^2}\int dk_{iz} |E_0(\omega_s + \omega_i(k_i) - \omega_p)|^2 \\ &\quad \times \cos^2\left\{[\omega_s + \omega_i(k_i)]\frac{T_p}{2}\right\} \\ &\quad \times \mathrm{sinc}^2\left\{\frac{L}{2}\left(k_p - k_{sz} - k_{iz} + \frac{\omega_s + \omega_i(k_i) - \omega_p}{u_p}\right)\right\}, \end{aligned} \tag{5}$$

where $\omega_i(k_i)$ is the dispersion dependence for the idler and $k_{ix} = -k_s \sin\theta_s$, with θ_s denoting the angle between $\mathbf{k}_s$ and the pump propagation direction (z axis). (The angle θ_s is also often referred to as the *angle of scattering* in the text in a sense that SPDC is often called parametric scattering.) The cosine modulation in Eq. (5) will not be averaged out by the integration over k_{iz} if the squared sinc function is much narrower than this modulation. In this case the squared sinc acts as a δ function in the integral; thus

$$R_s(\omega_s, \theta_s) \sim |E_0(\omega_s + \omega_i(k_i) - \omega_p)|^2 \cos^2\left\{[\omega_s + \omega_i(k_i)]\frac{T_p}{2}\right\}. \tag{6}$$

The R_s dependence on θ_s can easily be understood as follows. Since we are dealing with collinear degenerate SPDC, $\omega_i(k_i)$ can be expanded around $\omega_p/2$. Using the fact that the sinc-square function in Eq. (6) acts as a δ function and $\omega_s = \omega_p/2$, we obtain

$$\omega_i(k_i) \approx \frac{\omega_p}{2} + \frac{\theta_s^2 k_s}{2(u_i^{-1} - u_p^{-1})}\left(1 + \frac{k_s}{k_i(\omega_p/2)}\right), \tag{7}$$

where u_i is the group velocity of the idler inside the BBO crystal.

Clearly, Eq. (6) shows interference structure for the single counting rate of the signal detector. This modulation can be observed in several ways: (i) vary θ_s with fixed ω_s and T_p, (ii) vary T_p with fixed ω_s and θ_s, and (iii) vary ω_s with fixed T_p and θ_s. In this work, we only consider case (i). Case (ii) is considered in Ref. [13] and case (iii) is technically very difficult.

Let us now return to Eq. (5). The sinc-square function in the integral (5) is much narrower than the cosine modulation if

$$\frac{1}{d\omega_i/dk_{zi}} \gg \frac{T_p}{L}\frac{1}{\left(-1 + \frac{1}{u_p}d\omega_i/dk_{zi}\right)}. \tag{8}$$

For collinear SPDC, we can take approximately $d\omega_i/dk_{zi} = d\omega_i/dk_i \equiv u_i$. Hence, we obtain the following condition for observing the first-order interference in SPDC from a two-pulse pump:

$$Q \equiv \frac{L(u_p^{-1} - u_i^{-1})}{T_p} \gg 1. \tag{9}$$

Note that, although it is the signal that is being detected, Eq. (9) contains only the pump and the idler parameters. The other condition is obtained from the assumption we have used to get Eq. (6): the signal detector should select a sufficiently narrow frequency band and a sufficiently small solid angle (collinear degenerate SPDC). Indeed, the interference structure will be wiped out if Eq. (6) is integrated over a broadband of signal frequencies (ω_s) or angles of scattering (θ_s). Thus the requirement to the bandwidth of the signal detector is

$$\Delta\omega_s \ll \frac{\pi}{T_p}. \tag{10}$$

Equations (9) and (10) are the conditions for observing the first-order interference of SPDC in a two-pulse pump scheme.

What is the physical meaning of the above conditions? Considering only the signal photons, it would seem that the only condition for the first-order interference to take place is Eq. (10), which states that the filter inserted in front of the signal detector should have smaller bandwidth compared to the pump spectrum modulation. Indeed, if the signal photon wave packets are spread in time by more than T_p, the signal photons born of different pump pulses would seem indistin-

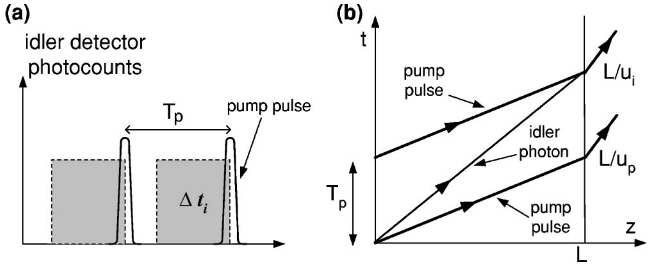

FIG. 2. (a) Photocounts from a broadband idler detector could be used to distinguish between signal photons generated from different pump pulses. Each idler photon comes after the corresponding pump pulse, with the time indeterminacy of $\Delta t_i = L(u_i^{-1} - u_p^{-1})$ (shown by a shaded region following the pump pulse). If $\Delta t_i < T_p$, signal photons from different pump pulses are distinguishable. (b) Feynman diagram illustrating the delay between the pump pulse and the idler photon inside the BBO crystal. If the crystal thickness is smaller than L shown in the figure, no interference can be observed.

guishable at first sight. The indistinguishability criterion, however, should be understood as indistinguishability *in principle. In principle*, we could equip our setup with an idler detector with a broadband filter and register photocounts from the idler detector. Then for each signal photon, detection of its twin idler photon is well localized in time with respect to the pump pulses (Fig.2), which could mean that we can always distinguish between a pair born of the first pulse and a pair born of the second pulse. Let us recall now that the BBO crystal has finite length L. Then a photocount in the idler detector can appear delayed from the corresponding pump pulse by any time $0 < t < \Delta t_i, \Delta t_i = L(u_i^{-1} - u_p^{-1})$; see Fig. 2(a). Idler photocounts from different pump pulses become indistinguishable if $\Delta t_i \gg T_p$, which is the second necessary condition for interference, Eq. (9).

In the experiment, the pump is the frequency-doubled laser pulse (400 nm) from the output of a mode-locked Ti:sapphire laser (800 nm). After frequency doubling, the pulse duration is 140 fsec, and the repetition rate is 90 MHz. The pump pulse is then fed into the polarization pulse splitter consisting of two Glan prisms (G) and a set of quartz rods QR placed between the prisms (Fig. 1). The axes of the Glan prisms are parallel to the pump polarization. The ''fast'' and ''slow'' axes of the quartz rods lie in the plane normal to the pump beam and are directed at 45° to the pump polarization. Due to the birefringence of the quartz rods, at the output of the polarization splitter each pump pulse is transformed into two pulses that have equal amplitudes but are delayed in time with respect to one another by $L_q(u_o^{-1} - u_e^{-1})$, where L_q is the total length of the quartz rods and u_o, u_e are group velocities of the ordinary and extraordinary waves in quartz at the pump wavelength (400 nm). SPDC is generated in a BBO crystal (thickness=3 mm) cut for collinear frequency-degenerate type-II phase matching. A prism (P) and a pinhole separate SPDC from the pump. A uv cutoff filter (F) reduces the pump fluorescent noise. A polarizing beam splitter (PBS) separates the signal from the idler, and the idler is discarded. The detector (EG&G SPCM-AQ-142) is placed at the focal plane of a lens ($f = 20$ cm), so that the transverse

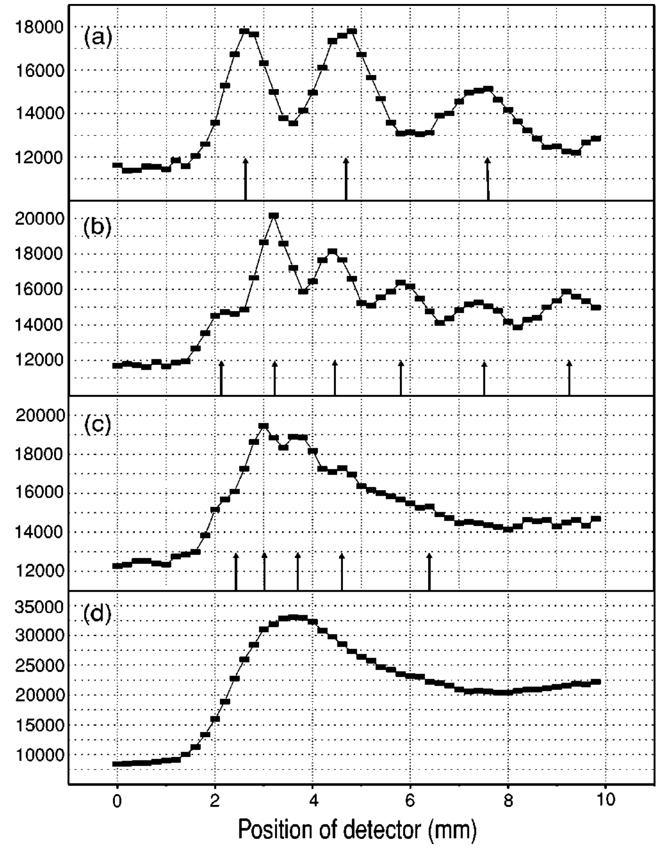

FIG. 3. Angular distribution ($\theta_s = x/f$, where x is the position of the detector and $f = 20$ cm) of the signal detector counts for three values of T_p: (a) 279 fsec, (b) 465 fsec, (c) 744 fsec. For all three plots, data accumulation time is 100 sec each. Arrows indicate the calculated positions of interference maxima. Plot (d) is data for a single pump pulse case. Data accumulation time is 30 sec. For all four plots, a 1-nm full width at half-maximum (FWHM) interference filter is used. The size of each data point corresponds to the uncertainty.

displacement of the detector is proportional to the angle of scattering, $x \sim f\theta_s$. In front of the detector, we place one of the three narrow-band filters (IF) with central wavelength $\lambda_s = 2\lambda_p = 800$ nm and bandwidths $\Delta\lambda_s = 1$, 3, and 10 nm, respectively, for different measurements. The intensity of the signal is measured as a function of the angle of scattering, $I_s(\theta_s)$. The angle is scanned by using a step motor (SM) that moves the detector in the focal plane of the lens. The parameters are bandwidth of the filter $\Delta\lambda_s$ and the time delay between the two pump pulses T_p, which is varied by using quartz rods with total length $L_q = 20$, 12.5, and 7.5 mm, corresponding to the delays $T_p = 744$, 465, and 279 fsec, respectively.

The fact that each pair of pulses is actually repeated at a rate of 90 MHz leads to a fine structure in the single-counting distribution (6), which is much narrower than the bandwidth of the filters we use in our experiment. Therefore, it is not observable.

In Fig. 3, the intensity is plotted versus the detector displacement for three delays T_p. All dependencies are obtained with the 1-nm interference filter. Note that in this case, condition (10) is satisfied for all three delays: $\pi/\Delta\omega_s \sim 2000$ fsec. However, the visibility is different in all three plots. The highest visibility of the interference pattern is observed for the smallest time interval between the pulses, 279 fsec, with $Q \sim 3$ [Fig. 3(a)]. For the intermediate delay, 465 fsec, the interference pattern is observed with lower visibility [Fig. 3(b)]. In this case, $Q \sim 2$. Evidently, Eq. (9) is not satisfied for the largest delay 744 fsec ($Q \sim 1$); therefore, in this case the interference structure completely vanishes [Fig. 3(c)]. For comparison, the angular spectrum of SPDC in the case of a single-pulse pump is shown in Fig. 3(d). In agreement with Eq. (6), the modulation period is larger for smaller delays; and the positions of the oscillation peaks are determined by the delay T_p introduced between the two pump pulses, in perfect agreement with the theoretical calculation [shown by arrows in Figs. 3(a)–3(c)]. However, the theory discussed above does not give an explicit description of the observed asymmetry of the angular spectral envelope. In this Rapid Communication, we only focus on the interference modulation; the shape of the angular spectrum envelope will be discussed elsewhere.

The spectral width of the filter, $\Delta\lambda_s$, has a strong influence on the interference pattern. Changing the filter bandwidth from 1 nm to 3 nm, we observed a considerable decrease of the visibility. At $\Delta\lambda_s = 10$ nm, no interference structure was observed, in agreement with condition (10).

There is an analogy between the first-order interference observed for SPDC generated from two spatially separated domains and for SPDC generated from two temporally separate pump pulses. Indeed, condition (9) ensures that the crystal is long enough so that an idler photon generated by the first pump pulse can meet the second pulse; see Fig. 2(b). In the case of spatially separated SPDC sources, first-order interference [6,8] is possible when idler waves propagate through both spatial domains where SPDC takes place [15]. Similarly to Ref. [8], where the effect has a simple explanation in terms of the pump angular spectrum, here it can be explained by the cosine modulation of the pump frequency spectrum. Condition (9) has the following spectral interpretation: the typical scale of the pump spectrum modulation should be much larger than the width of the idler spectrum, which is determined by the length of the crystal.

In conclusion, we have demonstrated the first-order interference of nonclassical light generated from two pump pulses well separated in time. The interference is explained by a quantum-mechanical calculation in terms of biphoton states. The interference pattern is observed in the angular distribution of the signal intensity. Interference is observed if the following condition is satisfied: the time indeterminacy of the delay between the idler photon and the corresponding pump pulse is much larger than the time interval between the pump pulses. From the spectral viewpoint, this condition means that the modulation of the pump spectrum, determined by the distance between the pulses, should be much larger than the width of the idler spectrum from a cw pump, determined by the crystal length. Thus the interference visibility is sensitive to the crystal length. It is also sensitive to the spectral width of the narrow-band filter used for the frequency selection of the signal photon.

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