Quantum interference and indistinguishability with femtosecond pulses

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The generation of entangled states of three or more photons through the nonlinear process of spontaneous parametric down-conversion requires the use of a femtosecond pump to provide an indistinguishability of photon pairs generated from different nonlinear crystals. We demonstrate experimentally that the quantum interference between two photons generated in each of the nonlinear crystals will degrade significantly as the duration of the femtosecond pump pulse becomes shorter than the coherence time of the signal and idler photons. Spectral postselection using narrow-band filters compensates this effect at the expense of the photon flux available for detection. [S1050-2947(97)50607-X]

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Entangled states of two or more quantum particles are at the heart of all major paradoxes associated with the interpretations of quantum mechanics [1,2]. The two-photon state generated in the nonlinear process of spontaneous parametric down-conversion (SPCD) has been an effective and convenient source of two-particle entanglement used in multiple tests of the Bell inequalities over the last decade [3]. A number of experimental techniques have been developed to manipulate significant quantum variables of the state. A particularly useful type of two-photon state that is entangled simultaneously in energy, space-time, and momentum was demonstrated recently by taking advantage of type-II SPCD [4]. The ultimate test of the nonlocal properties of quantum mechanics was proposed some time ago based on the use of multiple-particle entangled states [5]. The entangled states of two or more photons have also assumed an important place in the practical development of quantum cryptography techniques [6] and in the construction of basic elements of quantum teleportation to serve as a part of the quantum computing process [7].

The most convenient practical realization of such many-photon entangled states is the manipulation of the outcome of SPCD processes from several independent nonlinear crystals that are pumped by the same laser source. This technique relies on an important condition: The duration of the pump pulse must be much shorter than the coherence time of the signal and the idler photons generated in each SPDC crystal [8]. This condition establishes a mutual indistinguishability of multiple pairs generated from independent crystals and creates the basis for the coherent manipulation of multiple probability amplitudes. This usually requires the use of a femtosecond pump pulse. In the spectral domain this means that the spectrum of the pump pulse must be wider than the spectral widths of the signal and the idler photons that are defined by the natural phase matching (plane-wave model for the pump) in nonlinear parametric processes.

In this paper we demonstrate that the enforcement of the above condition results in a strong decoherence of the output signal from each of the down-conversion crystals. This leads to the loss of stable phase relationships between the optical field amplitudes in the signal and idler modes and destroys the high visibility quantum interference, even with a single crystal. We report on the experimental demonstration of the degradation of two-photon quantum interference in SPDC with a femtosecond pump source. The only technique available so far to counter this problem is the spectral postselection of the entangled photons by the placement of narrow-band interference filters in front of detectors [12]. While restoring the high visibility quantum interference, this unfortunately reduces the number of photons reaching the detector. We evaluate the impact of the narrow-band filters on the results of such measurements.

The basic schematic of our experimental setup for the study of decoherence effects in SPDC with a femtosecond pump is illustrated in Fig. 1. The pulsed radiation generated from the Ti:sapphire femtosecond laser (Coherent Mira 900) was selected to have a central wavelength of 795 nm. After the conversion to a second harmonic, the 155-fs duration output pulse with central wavelength 397.5 nm is directed towards the down-conversion nonlinear crystal. A two-photon quantum state is generated in the collinear configuration type-II SPDC. In our experiment we use a BBO (β-BaB_2O_4) nonlinear crystal oriented at 42.3° relative to

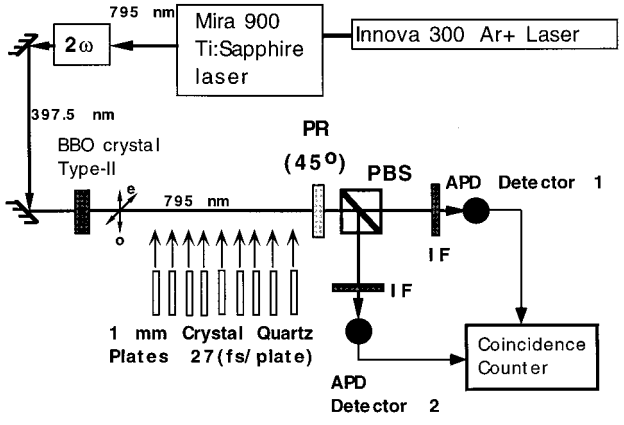

FIG. 1. Schematic of the experimental setup.

the optical axes. It is technically rather difficult to vary the pulsewidth of a femtosecond laser over a wide range. As an alternative we utilized three pieces of nonlinear crystal with thicknesses of 3, 1.5, and 0.56 mm to provide three options for the value of the ratio of the signal coherence time to the pump pulse duration: 2.7, 1.35, and 0.45, respectively. It is known from the theory of parametric processes in nonlinear optics [9–11] that the coherence time of the signal or idler wave can be estimated from the spectral width of the natural phase matching. In the first order this will be defined by the dispersion of the group velocities for the ordinary (o) and extraordinary (e) polarized waves in the medium

$$t_{\mathrm{coh}}=\left(\frac{1}{u_e}-\frac{1}{u_o}\right)L \tag{1}$$

where L is the length of the nonlinear crystal. Three samples of BBO crystal in our experiment create a down-converted optical field with a central wavelength of 795 nm and a coherence time of 420, 210, and 70 fs, respectively.

A simple correlation technique [13,14] was used to prepare an entangled configuration of two-photon probability amplitudes and to detect a high visibility quantum interference modulation of coincidence counts. This method is based on the use of a polarizing beam splitter (PBS) and a 45° polarization rotator in front of the beam splitter. This design ensures the equal probability for each of the original ordinary (vertical) and extraordinary (horizontal) components of a two-photon state to reach both photon-counting detectors D_1 and D_2. We used a pair of EG&G passively quenched avalanche photodiodes (APDs) in the Giger mode to detect single photon events. The probability of coincidence detection (number of coincidence counts per second) is a function of the relative optical path delay τ between the ordinary and extraordinary components of the original state. We introduce a variable optical delay line made of thin (1-mm) plates of crystal quartz in front of the beam splitter in order to detect a change in the degree of quantum interference (visibility of coincidence probability modulation) as a function of the quantum amplitudes' indistinguishability (their overlap in longitudinal space-time) [14].

The experimental data from three runs with different thicknesses of nonlinear crystals are presented in Fig. 2. These results clearly demonstrate the degradation of quantum interference when the spectrum of the pump pulse becomes wider than the natural bandwidth of phase matching. The experimental data were obtained with relatively broad (6-nm full width at half maximum) interference filters in front of the detectors to reduce the influence of ambient noise on the experimental results. The equivalent coherence time of the light passing through such a filter at 795 nm is 350 fs. This time is greater than the pump duration and the overall influence of our noise cutoff filters on the effect of visibility reduction is very small (see Fig. 3). The convolution of the filter spectral profile with the natural phase matching and with the spectrum of the pump will define the correlation function measured in our experiment for each of the crystal lengths. Each step in the delay line corresponds to an optical delay of 9 μm (or delay time of 27 fs) in crystal quartz between the ordinary and extraordinary waves. The horizontal shift of the absolute minima of the dip also depends on the relation between the pump spectrum and the natural width of the phase matching in the nonlinear crystal. This effect comes naturally from our theory and will be discussed in detail elsewhere.

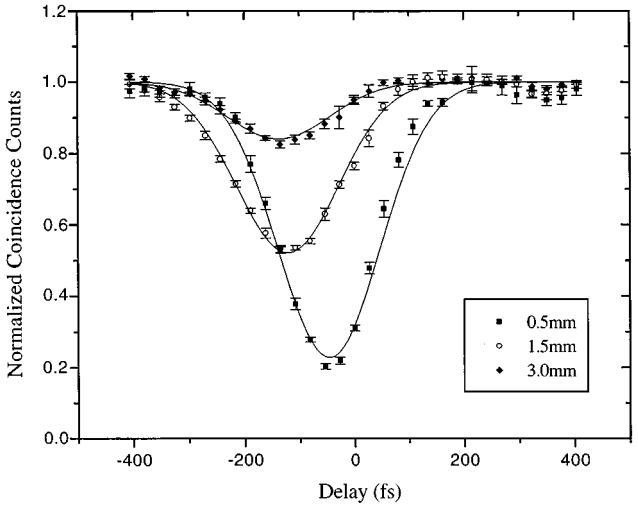

FIG. 2. Quantum interference measurement with three nonlinear crystals of different length 0.56, 1.5, and 3 mm.

To develop a simple description of the demonstrated effect we will ignore the components of the wave vectors perpendicular to the pump beam and will drop the vector notation for the wave vector. Then the wave function at the output surface of the crystal may be written as

$$|\Psi\rangle=|0\rangle+\sum_{k_p}A(k_p)\sum_{k,k'}F_{k,k'}\hat{a}^{\dagger}_{ok}\hat{a}^{\dagger}_{ek'}|0\rangle. \tag{2}$$

The coefficient $F_{k,k'}$ is given by [10,15]

$$F_{k,k'}=\Gamma_{k,k'}L\,\delta(\omega_{ok}+\omega_{ek'}-\omega_p)h(L\Delta_{k,k'}), \tag{3}$$

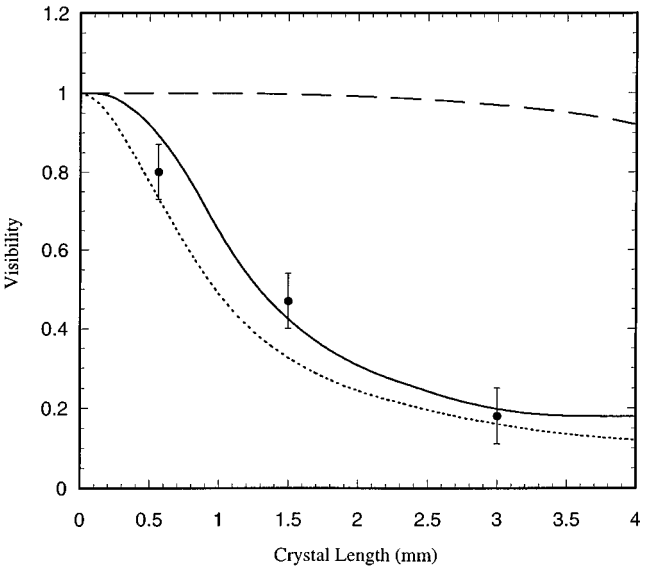

FIG. 3. Comparison of quantum interference visiblity observed in the experiment with the results of theoretical prediction using 6-nm interference filters (solid line). Theoretical prediction with 0.6-nm interference filters (dashed line) and without any filters inserted (dotted line).

where $\Gamma_{k,k'}$ is the parametric gain index for perfect phase matching (it is a sufficiently slowly varying parameter over the bandwidth of the down-conversion process so that it may be taken as a constant). The δ function represents the frequency phase-matching condition, and the h function, where $h(x)=[1-e^{-ix}]/ix$ and $\Delta_{k,k'}=k_p-k'-k$, determines the natural spectral width of the two-photon state in the nonlinear crystal. If the spectral bandwidth of the pump, treated classically as a plane wave, is small compared to its center (carrier) frequency Ω_p, the electric field of the pump is represented as $E_p(t)=E_{\rm env}(t)\exp(i\Omega_p t)$, where $E_{\rm env}(t)$ is the complex pulse envelope and $A_{\rm env}(k_p)$ is its Fourier transform [16,17].

The coincidence counting rate between detectors D_1 and D_2 is defined by

$$R_c=\lim_{T\to+\infty}\frac{1}{T}\int_{-T/2}^{T/2}dT_1 \times\int_{-T/2}^{T/2}dT_2\langle\Psi|\hat{E}_1^{(-)}\hat{E}_2^{(-)}\hat{E}_2^{(+)}\hat{E}_1^{(+)}|\Psi\rangle S(T_1-T_2), \tag{4}$$

where $S(t)$ is the coincidence window function that is defined such that $S=1$ for $|t|<t_{\rm coinc}$ and goes to zero rapidly for $|t|>t_{\rm coinc}$. In our experiments we have a train of pump pulses, so that the coincidence rate can be defined as

$$R_c=f_p\int_{-\infty}^{+\infty}dT_1\int_{-\infty}^{+\infty}dT_2\langle\Psi|\hat{E}_1^{(-)}\hat{E}_2^{(-)}\hat{E}_2^{(+)}\hat{E}_1^{(+)}|\Psi\rangle, \tag{5}$$

if $f_p^{-1}>t_{\rm coinc}\gg\delta t_{\rm pulse}$, where f_p is the repetition rate of the pulsed laser and $\delta t_{\rm pulse}$ is the duration time of the pulse.

The two-photon amplitude $A(t_1,t_2)$ is defined as [15]

$$|A(t_1,t_2)|^2=\langle\Psi|\hat{E}_1^{(-)}\hat{E}_2^{(-)}\hat{E}_2^{(+)}\hat{E}_1^{(+)}|\Omega\rangle =|\langle 0|\hat{E}_2^{(+)}\hat{E}_1^{(+)}|\Psi\rangle|^2, \tag{6}$$

where, for simplicity, the calculation is in the Heisenberg picture in which the state vector is the state at the output face of the crystal. Assuming point detectors without filters, the quantized fields are

$$\hat{E}_1^{(+)}=\sum_k E_{jk}\hat{a}_{jk}e^{-i\omega_k t}. \tag{7}$$

If the polarizing beam splitter transmits the o ray into the detector D_1 and reflects the e ray into D_2, we have

$$A(t_1,t_2)=\sum_{k_p}A(k_p)\sum_{k_1,k_2}E_{k_1}E_{k_2}e^{-i\omega_{k_1}t_1-i\omega_{k_2}t_2}\langle 0|\hat{a}_{ok_1}\hat{a}_{ek_2}|\Psi\rangle =W_0e^{-i(\Omega_d/2)(t_1-t_2)}\Pi(t_1-t_2)e^{-i(\Omega_p/2)(t_1+t_2)}E_{\rm env}\left(\frac{t_1+t_2}{2}+\frac{\Lambda}{D}(t_1-t_2)\right) \tag{8}$$

where $\Omega_d=\Omega_o-\Omega_e$ (we have perfect phase matching at the ordinary and extraordinary angular frequencies Ω_o and Ω_e), $D=1/u_o-1/u_e$, and $\Lambda=1/u_p-\frac{1}{2}(1/u_o+1/u_e)$. The function $\Pi(x)$ describes the accumulation of optical delay t_1-t_2 between the two photons in the pair (which is proportional to the length of their travel inside the crystal). The Π function also illustrates the random probability distribution for two-photon creation at any point inside the nonlinear crystal during the coherent nonlinear interaction. A detailed description of this function can be found in [15]. The rectangular shape of $\Pi(x)$ in the time domain is responsible for the observation of a triangular shape intensity correlation function in the coincidence measurement [14]. All of the slowly varying quantities and constants have been absorbed in W_0. It is known that the entangled two-photon state does not factor into a product of a function of t_1 and a function of t_2, but it can be represented as a product of two linear combinations that depend on t_1-t_2 and t_1+t_2 in the case of a monochromatic continuous-wave pump [15]. Now, from Eq. (8) we conclude that if the pump is pulsed and has the shape of a wave packet that is comparable in spectral width with the natural bandwidth of phase matching in the nonlinear crystal of length L, then the two-photon wave function does not factor into a simple product of functions of t_1-t_2 and t_1+t_2. This is because the pair cannot be created with equal probability (i.e., randomly) throughout the nonlinear crystal as in the cw regime. The pulsed pump introduces a ''knowledge in principle'' of the time creation of the pair. For example, tracking the pump pulse position inside the crystal, we could discriminate between pairs created at the input surface of the crystal and pairs created at the output surface of the crystal. The rectangular function $\Pi(t_1-t_2)$ describes the fact that the pair is created at the same time and at the same point inside the crystal, but the wave packets travel through the crystal with different group velocities, so the complex shape function $E_{\rm env}((t_1+t_2)/2+\Lambda/D(t_1-t_2))$ describes this ''knowledge in principle'' of the creation time (or the creation point inside the crystal).

If we consider that narrow-band frequency filters centered at perfect phase matching were placed in front of the detectors then the fields must be written as

$$\hat{E}_1^{(+)}=\sum_k f_j(\omega_k-\Omega_j)E_{jk}\hat{a}_{jk}e^{-i\omega_k t}, \tag{9}$$

and the Π-function now also depends on the t_1+t_2 time parameter $[\Pi_f(t_1-t_2,t_1+t_2)]$. Physically, we can interpret this procedure of the postselection of spectral amplitudes by

narrow-band frequency filtering as a method to significantly erase the ''knowledge in principle'' introduced by the femtosecond pump radiation.

The final effective two-photon wave function can be presented in a familiar form:

$$\Psi(t_1,t_2)=[(\boldsymbol{\epsilon}_1\cdot\boldsymbol{\epsilon}_0)(\boldsymbol{\epsilon}_2\cdot\boldsymbol{\epsilon}_e)A(t_{1o},t_{2e}) + (\boldsymbol{\epsilon}_1\cdot\boldsymbol{\epsilon}_e)(\boldsymbol{\epsilon}_2\cdot\boldsymbol{\epsilon}_o)A(t_{2o},t_{1e})] \quad (10)$$

where $\boldsymbol{\epsilon}_j$ represents the unit vectors parallel to the polarization axes of the SPDC crystal and the polarization beam splitter. Considering an ideal polarization beam splitter and a 45° rotation of the SPDC crystal polarization by a polarization rotator, as well as the relative optical delay τ between the *o* and *e* rays introduced by quartz plates, we obtain a final expression for the coincidence counting rate:

$$R_c=R_0[1-\rho(\tau)] \quad (11)$$

with

$$R_0=\int_{-\infty}^{+\infty}dT\int_{-\infty}^{+\infty}dt|\Pi_f(t,T)E_{\mathrm{env}}(t,T)|^2, \quad (12)$$

$$\rho(\tau)=\frac{1}{R_0}\int_{-\infty}^{+\infty}dT\int_{-\infty}^{+\infty}dt\ \mathrm{Re}\{\Pi_f^*(t+\tau,T)\Pi_f(-t+\tau,T) \times E_{\mathrm{env}}^*(t+\tau,T)E_{\mathrm{env}}(-t+\tau,T)\}, \quad (13)$$

and $T=t_1+t_2$, $t=t_1-t_2$.

The theoretical prediction for the quantum interference visibility as a function of the length of the BBO nonlinear crystal in the case of 155-fs pulsed pump radiation is illustrated in Fig. 3 with a solid line to match the data from our experiment. The visibility was calculated using Eq. (13) and assuming the use of 6-nm bandwidth filters to match our experiment. The dotted line corresponds to the predicted visibility reduction when no additional spectral filtering is introduced. The instability of the femtosecond pulse duration over the time of the measurements with each crystal was the main contribution to the experimental error. The dashed line illustrates a theoretical possibility of recovering a quantum interference visibility with a spectral postselection procedure using very-narrow-band 0.6-nm interference filters in front of the detectors. Assuming Gaussian filters, this will decrease the intensity of two-photon states by at least one order of magnitude. We expect even greater losses if the real spectral shape of a single-cavity subnanometer interference filter is considered.

In conclusion, we have experimentally demonstrated the effect of quantum interference degradation during the SPDC process when the duration of the pump pulse becomes shorter than the coherence time of the signal radiation. The pulsed pump introduces a ''knowledge in principle'' of the creation time of the photon pair. The high visibility of quantum interference can be recovered to some degree by performing the postselection of spectral amplitudes using very-narrow-band interference filters in front of the photon-counting detectors. However, this results in a significant reduction of the number of available two-photon states. These results have important implications in the area of practical two-photon cryptography and multiple-particle quantum interferometry, including quantum teleportation.

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