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Spectral distinguishability in ultrafast parametric down-conversion

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We report a fourth-order interference experiment in which pairs of photons produced in parametric down-conversion pumped by short optical pulses interfere in a Hong-Ou-Mandel interferometer. The visibility of the interference pattern is reduced for larger pump bandwidths. This effect can be understood in terms of the spectral distinguishability of the photon pairs. The interference can be restored by blocking the distinguishing information with a spectral filter. [S1050-2947(98)50704-4]

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Indistinguishability of possible pathways leading to the same outcome is an essential condition for quantum interference [1]. A useful tool for the study of this relationship is the Hong-Ou-Mandel interferometer [2], in which two photons produced in the process of parametric down-conversion are brought together at a beam splitter. In a recent experiment it was shown that two-photon interference can occur even when the two photons do not reach the beam splitter simultaneously [3], thus demonstrating that such interference should be regarded as the interference of the amplitudes for the possible two-photon paths, rather than as the interference of the two photons themselves. Experiments such as this can be understood in terms of the indistinguishability of the processes leading to a coincidence count. If ''which process'' information is present in a system, the interfering processes become distinguishable and the visibility of the interference pattern is reduced. The distinguishing information may be of any type. In one experiment, for example, the information is contained in the orthogonal polarizations of the photons in a Hong-Ou-Mandel interferometer [4]. This information makes it possible to identify the alternate Feynman paths and the interference is observed only when this information is blocked. The arrival times of the two photons at the detectors may also produce distinguishing information. A displacement of the beam splitter in the Hong-Ou-Mandel interferometer, for example, causes one of the photons to reach the beam splitter before the other. Complete interference occurs, however, only when there is absolutely no distinguishing time-of-flight information. A third type of information available in the down-conversion process is spectral information. This information is not present in the experiments described above because, as a consequence of the very narrow spectrum of the cw pump field, the two photons have identical spectra.

If the down-conversion process is pumped by a train of short optical pulses, with a correspondingly large bandwidth, the spectral correlation of the down-converted fields becomes more complex [5]. In certain cases, this additional complexity may introduce distinguishing information, which can degrade interference not only in a Hong-Ou-Mandel interferometer, but also in interference experiments involving photons created in independent down-conversion crystals. The latter is of interest in the context of the multiparticle interference experiment recently proposed by Greenberger, Horne, and Zeilinger [6]. The most practical technique proposed for the generation of the multiparticle entangled states required for such an experiment involves (nearly) simultaneous emission of correlated photon pairs from independent down-conversion crystals [7,8]. This can be accomplished only if the coherence time of the pump is on the order of the coherence times of the down-converted photons [9], a condition that is satisfied by an ultrafast pump source.

In this paper we describe a fourth-order interference experiment involving the photon pairs emitted from a type-II down-conversion crystal pumped by an ultrafast source. The emitted pairs, in addition to being distinguishable by their orthogonal polarizations, can also be identified with some degree of certainty by their joint spectra. We study the effect of this spectral distinguishability by interfering the photon pairs in a Hong-Ou-Mandel apparatus, in which the photons are rendered indistinguishable with regard to polarization. The degree of spectral distinguishability, which depends both on crystal parameters and on the pump bandwidth, is then manifested in the visibility of the fourth-order interference between alternate two-photon detection processes. In the experiment described below, interference patterns are recorded for two different pump bandwidths, while the crystal parameters are treated as experimental constants. We find that the visibility is reduced for the larger pump bandwidth, but can be restored by placing a spectral filter in front of the detectors, effectively blocking the distinguishing ''which process'' information. Of course, this is done at the expense of a lower count rate.

The experimental apparatus is shown schematically in Fig. 1. The frequency-doubled output of a mode-locked Ti:sapphire laser is used to pump a 0.8-mm BBO crystal oriented for collinear type-II down-conversion. The first of the two alternate pump fields consists of a train of pulses centered at 405 nm with a bandwidth of 3 nm. A nondispersive sequence of prisms serves as a filter to block the funda-

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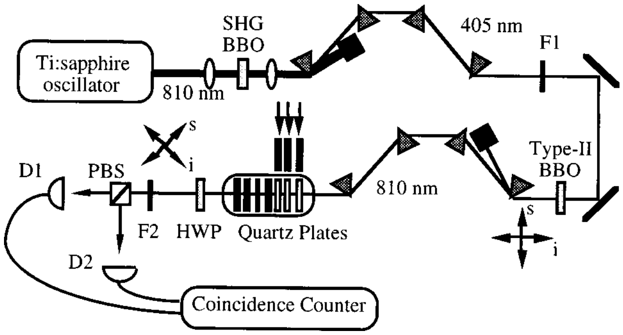

FIG. 1. Schematic of the experimental setup. SHG is a frequency-doubling crystal, F1 and F2 are interference filters, HWP is a half-wave plate oriented to rotate the polarizations of the down-converted photons by 45°, PBS is a polarization beam splitter, and D1 and D2 are avalanche photodiodes.

mental wavelengths. The second pump field is produced by passing this beam through a spectral filter with a bandwidth of 0.8 nm. The output photons are orthogonally polarized and, after separation of the residual pump radiation via a second nondispersive prism sequence, travel a common path through a Hong-Ou-Mandel interferometer. A pair of photon-counting avalanche photodiodes monitor the two outputs of a polarization beam splitter. A waveplate is placed in front of the beam splitter to rotate the polarizations of the down-converted photons to $\pm 45°$ with respect to the axes of the polarization beam splitter. This combination of waveplate and polarization beam splitter behaves as a 50/50 beam splitter for each of the incident photons. The coincidence rate is measured as a function of relative delay between the paths taken by the orthogonally polarized photons. This delay is adjusted by inserting a number of crystal quartz plates in the beam. The quartz is oriented so that its fast and slow axes are aligned with the polarizations of the two photons. In the first experiment, broad spectral filters are placed before the detectors and are used only to reduce spurious background counts. These filters are replaced by narrow-band filters in the second experiment. Their effect is discussed below.

The state vector for the signal and idler photons after the down-conversion crystal may be written as [5]

$$|\Psi\rangle = \int \int d\omega_s d\omega_i \alpha(\omega_s + \omega_i) \times \Phi(\omega_s, \omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) |\text{vac}\rangle. \tag{1}$$

This is a continuous superposition of two-photon states in which the signal and idler photons have frequencies ω_s and ω_i, respectively. The probability amplitude for each pair of frequencies is the product of an energy function $\alpha(\omega_s + \omega_i)$ and a phase-matching function $\Phi(\omega_s, \omega_i)$. The energy function $\alpha(\omega_s + \omega_i)$ is simply the Fourier transform of the time-dependent part of the classical pump field. Its presence ensures that a given pair of frequencies may be found in the signal and idler fields only if they sum to a frequency found in the pump. While the energy function determines the pump frequencies available for down-conversion, the phase-matching function determines how these energies may be distributed to the down-converted fields.

A pair of photons emitted in the down-conversion process is said to be spectrally distinguishable if knowledge of both frequencies allows the photons to be identified as either the signal (ordinary polarization) or idler (extraordinary polarization). It should be stressed that it is the *combination* of frequencies that provides the distinguishing information. For a given pair of frequencies, two possibilities exist: either the signal was found with one frequency and the idler with the other, or vice versa. The distinguishability of these two situations depends on the form of the probability amplitude in Eq. (1). In particular, the two cases are indistinguishable, or equally likely, if the value of the probability amplitude remains unchanged when the order of the two frequency arguments is reversed. The energy function, which depends on the *sum* frequency, does possess this symmetry; therefore, the two possible pair configurations will be spectrally indistinguishable if the phase-matching function is also symmetric, that is, if $\Phi(\omega_1, \omega_2) = \Phi(\omega_2, \omega_1)$. In general, however, the phase-matching function does not possess this symmetry. For a type-II crystal of length L oriented for phase-matched degenerate down-conversion of frequency $2\overline{\omega}$, the phase-matching function may be approximated by [5]

$$\Phi(\omega_s, \omega_i) = \frac{\sin\{[(\omega_s - \overline{\omega})(k_s' - k_p') + (\omega_i - \overline{\omega})(k_i' - k_p')]L\}}{[(\omega_s - \overline{\omega})(k_s' - k_p') + (\omega_i - \overline{\omega})(k_i' - k_p')]L}, \tag{2}$$

where $k_p' = \partial k_p(\omega)/\partial\omega|_{\omega = 2\overline{\omega}}$ and $k_j' = \partial k_j(\omega)/\partial\omega|_{\omega = \overline{\omega}}$ for $(j = s, i)$. This expression is not symmetric with respect to its two frequency arguments because the two axes of the down-conversion crystal have different dispersive properties and, therefore, $k_s' \neq k_i'$ [10].

The effect of the above asymmetry is completely obscured in the case of cw-pumped degenerate down-conversion, as can be seen by examining the energy function $\alpha(\omega_s + \omega_i)$. For a single-frequency cw pump of frequency $2\overline{\omega}$, the energy function takes the form $\alpha(\omega_s + \omega_i) = \delta(\omega_s + \omega_i - 2\overline{\omega})$. If, as assumed above, the crystal is aligned for perfect phase matching for degenerate down-conversion, then the most probable frequency for each of the down-converted fields is $\overline{\omega}$. Other pairs of frequencies are generated, as well, with each pair satisfying the constraint imposed by the delta energy function, so that if a measurement of the frequency of the signal photon yields the result $\omega_s = \overline{\omega} + \delta\omega$, then the idler photon will have the frequency $\omega_i = \overline{\omega} - \delta\omega$. The form of the phase matching function in Eq. (2) is such that this result is just as likely as the result $\omega_i = \overline{\omega} - \delta\omega$ and $\omega_s = \overline{\omega} + \delta\omega$. In this case, therefore, frequency measurements alone would yield no information about the identities (signal or idler) of the detected photons and each pair may be considered spectrally indistinguishable.

If the process is instead pumped by a monochromatic field of frequency $2\overline{\omega} + \delta\omega_p$, then the energy function has the form $\alpha(\omega_s + \omega_i) = \delta(\omega_s + \omega_i - 2\overline{\omega} - \delta\omega_p)$. In this case, which is phase matched for *non*degenerate down-conversion, the most probable frequencies for the signal and idler are those which simultaneously zero the argument of the sinc function in Eq. (2), and satisfy the δ function constraint. That is,

$$\omega_s=\overline{\omega}+\delta\omega_p\left(\frac{k_i'-k_p'}{k_i'-k_s'}\right),\quad \omega_i=\overline{\omega}-\delta\omega_p\left(\frac{k_s'-k_p'}{k_i'-k_s'}\right). \tag{3}$$

Thus, the center frequencies of the down-converted fields are shifted away from degeneracy. The magnitudes of these shifts depend linearly on $\delta\omega_p$, but because $k_s'\neq k_i'$, the shift is larger for one photon than for the other. This has the effect of making the photon pair spectrally distinguishable, increasingly so for larger values of $\delta\omega_p$. Thus, while it is the asymmetry of the phase-matching function that ultimately leads to the spectral distinguishability of the photon pairs, the role played by the energy function is critical, since it determines the range of $\delta\omega_p$.

For the experimental apparatus in Fig. 1, the pump beam is composed of a train of short optical pulses that can be thought of as a superposition of a large number of single-frequency fields. The above argument follows for each spectral component of the pump and, therefore, each of these pump frequencies contributes a different amount of spectral distinguishability. If the pump spectrum is narrow in relation to the width of the phase-matching function, then none of the pump frequencies differs substantially from the center frequency $2\overline{\omega}$, and the degree of distinguishability is low. Pump fields with larger spectra include a larger proportion of frequencies far from the center frequency $2\overline{\omega}$ and the photon pairs are more distinguishable. Thus, while the asymmetry of the phase-matching function is essential to the distinguishability of the signal and idler, it is the size of the pump bandwidth that affects the *degree* of distinguishability [5].

As stated earlier, the signal and idler photons are initially distinguished by their polarizations, as well as by their joint spectral properties. However, after passing through the waveplate and the polarization beam splitter in the Hong-Ou-Mandel interferometer, the photons become polarization entangled, so that they can no longer be identified by polarization measurements alone. In this case, the visibility of the fourth-order interference is determined solely by the spectral distinguishability of the down-converted pairs. The degree of interference between alternate two-photon detection paths is seen in the coincidence counting rate

$$R_c(\delta\tau)=\frac{1}{T}\int\int_0^T dt_1dt_2 \times\langle\Psi|\hat{E}_1^{(-)}(t_1)\hat{E}_2^{(-)}(t_2)\hat{E}_2^{(+)}(t_2)\hat{E}_1^{(+)}(t_1)|\Psi\rangle. \tag{4}$$

Here, $\delta\tau$ is the relative polarization delay, T is the coincidence resolving time, and $\hat{E}_{1,2}^{(+)}(t)=(1/\sqrt{2})[\hat{E}_s^{(+)}(t)\pm\hat{E}_i^{(+)}(t+\delta\tau)]$ are the positive frequency pairs of the electric-field operators at detectors 1,2. The signal and idler field operators, in turn, may be written as $\hat{E}_{s,i}^{(+)}(t)\propto\int d\omega\hat{a}_{s,i}(\omega)e^{-i\omega t}$. When the field operators are expressed in this manner, and the state vector presented in Eq. (1) is inserted into Eq. (4), the result

$$R_c(\delta\tau)\propto\int\int d\omega_s d\omega_i|\alpha(\omega_s+\omega_i)|^2[|\Phi(\omega_s,\omega_i)|^2 -\Phi(\omega_s,\omega_i)\Phi^*(\omega_i,\omega_s)e^{-i(\omega_i-\omega_s)\delta\tau}] \tag{5}$$

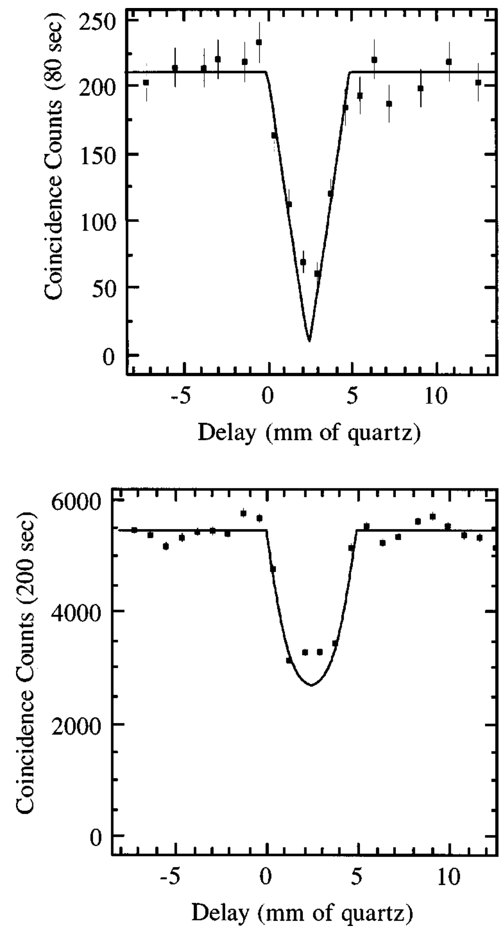

FIG. 2. Interference dip for two different pump bandwidths. With filter F1 in place (a), the pump bandwidth is 0.8 nm. With no filter (b), the bandwidth is 3 nm. The solid lines are fits to theory.

is obtained [5]. The first term is an integration of the total two-photon probability density distribution over all frequencies, and represents the total ''background'' rate of coincidence detection independent of $\delta\tau$. The second term, which contributes only for small values of $\delta\tau$, is the source of the coincidence ''dip'' observed in traditional experiments of the Hong-Ou-Mandel type. For $\delta\tau=0$, the coincidence rate reaches zero if $|\Phi(\omega_s,\omega_i)|^2=\Phi(\omega_s,\omega_i)\Phi^*(\omega_i,\omega_s)$ over the entire range of frequencies that contribute to the integral in Eq. (5). As discussed above, this range is set by the bandwidth of the pump spectrum specified by $\alpha(\omega_s+\omega_i)$.

The calculated results for the two pump bandwidths used in the experiment are plotted in Fig. 2, along with the experimental results. The theoretical curves [5] were generated as best fits to the data, with detection efficiency as the only free parameter, and assuming Gaussian shapes for the pump spectra. The recorded count rates are somewhat higher than expected near the centers of the dips. This discrepancy can be attributed to misalignment and to imperfect polarization components. The minima in the two interference patterns occur at the same relative delay, but the visibility is different for the two pump bandwidths. The curve obtained with the filtered pump, shown in Fig. 2(a), has a high degree of visibility, and comes close to the triangle shape reported for the interference pattern produced by the photon pairs generated in cw-pumped type-II down-conversion [10]. When the

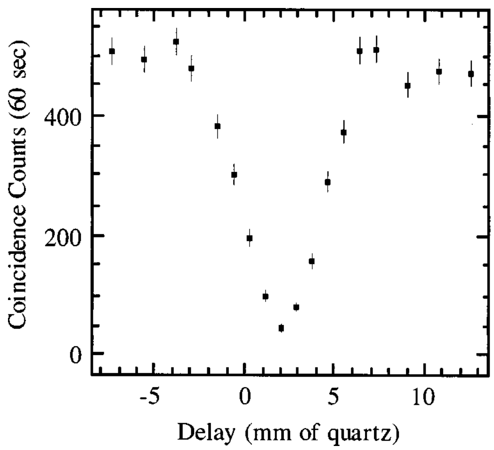

FIG. 3. Interference dip with filter F2 in place. As in Fig. 2(b), the pump bandwidth is 3 nm.

broadband pump (no filter) is used, the two-photon detection processes possess a higher degree of distinguishability, yielding an interference curve with diminished visibility, as shown in Fig. 2(b).

These data show that the visibility of the interference may be improved by limiting the pump bandwidth, but this obviously results in a longer pump coherence time. Another method is to allow only the indistinguishable photon pairs to reach the detectors. This can be accomplished by passing the photons through a spectral filter. For the data shown in Fig. 3, the large-bandwidth filter F2 was replaced with a spectral filter having a bandwidth of 10 nm, which is smaller than the bandwidths of the individual photons, thereby increasing their coherence times. However, the two-photon coherence time [11], which is determined by the crystal length and the difference in group velocities for the two down-converted fields, is relatively unchanged. The result is that the visibility of the interference pattern is significantly improved, while the ''width'' of the dip is preserved. The use of spectral filters is not uncommon in this type of experiment [2,11], in which a coincidence rate is measured for different values of delay. These experiments differ from the present one, though, because the filters are not, in principle, essential for high visibility in the cw case. They are helpful in a practical sense, in that they improve stability by increasing the coherence times of the photons, but the minimum coincidence rate is predicted to reach zero, even for the case of no spectral filtering. As noted above, this is a consequence of pumping the down-conversion process with a monochromatic field. The photon pairs produced in broadband-pumped down-conversion, on the other hand, are spectrally distinguishable, and so the minimum coincident rate is never zero. The role of the filter in the present application is to prevent the distinguishable pairs from reaching the detectors, thus improving the visibility. As discussed earlier in this paper, the photon pairs originating from the down-conversion of a pump photon with a frequency far from the center frequency $2\overline{\omega}$ are more readily identifiable as signal or idler than those originating from a pump photon with a frequency near the center frequency. Inspection of the relations in Eq. (2) show that these more distinguishable pairs are also those that have frequencies far from the center frequency $\overline{\omega}$, and that are therefore blocked by the narrow spectral filter.

We have experimentally demonstrated that the visibility in fourth-order interference of photons produced in type-II parametric down-conversion is reduced when the process is pumped by a short optical pulse. This effect can be understood in terms of the spectral distinguishability of the photon pairs. Shorter pulses, with larger bandwidths, produce a larger proportion of pairs whose ''which photon'' configurations are knowable in principle, thereby reducing the visibility. We have also shown that the interference can be recovered by passing the photons through a spectral filter. The filter effectively blocks the distinguishing information. This work helps to provide a better understanding of the dynamics of ultrafast down-conversion, thus making multiparticle interferometry more feasible.

Note added. Since completing this work, the authors learned of a recently published paper describing a similar experiment [12].

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