Joint Temporal Density Measurements for Two-Photon State Characterization

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We demonstrate a technique for characterizing two-photon quantum states based on joint temporal correlation measurements using time-resolved single-photon detection by femtosecond up-conversion. We measure for the first time the joint temporal density of a two-photon entangled state, showing clearly the time anticorrelation of the coincident-frequency entangled photon pair generated by ultrafast spontaneous parametric down-conversion under extended phase-matching conditions. The new technique enables us to manipulate the frequency entanglement by varying the down-conversion pump bandwidth to produce a nearly unentangled two-photon state that is expected to yield a heralded single-photon state with a purity of 0.88. The time-domain correlation technique complements existing frequency-domain measurement methods for a more complete characterization of photonic entanglement.

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Spontaneous parametric down-conversion (SPDC) is a powerful method for generating two-photon states for quantum information processing (QIP). The joint quantum state can be engineered for specific QIP applications by tailoring its polarization, momentum, and spectral degrees of freedom. Ultrafast-pumped SPDC is of great interest because a well-defined time of emission is desirable in clocked applications such as linear optics quantum computing [1]. In ultrafast SPDC, spectral engineering of the two-photon state can be accomplished by manipulating the crystal phase-matching function and the pump spectral amplitude [2,3] to yield unique forms of two-photon frequency entanglement. For example, coincident-frequency entanglement with strong positive correlation between signal and idler emission frequencies can be used to improve time-of-flight measurements beyond the standard quantum limit [4,5]. On the other hand, one can utilize a two-photon state with negligible spectral correlations to implement a heralded source of pure-state single photons [6,7].

Characterizing the spectral correlations of a two-photon state can be done by measuring the joint spectral density (JSD) profile with tunable narrow band filters [6–8]. Hong-Ou-Mandel (HOM) quantum interference [9] is also useful for quantifying the two-photon coherence bandwidth and the indistinguishability of the photon pair. However, the two measurements do not give the whole picture of the two-photon state. Both measurements are insensitive to the spectral phase and therefore cannot capture the time-domain dynamics unless the joint state is known to be transform limited. Moreover, JSD measurements with low detector efficiency or high detector noise can be challenging due to long acquisition times and low signal-to-noise ratios. Frequency-domain techniques for estimating the spectral phase exist, but they are not simple to implement in practice [10].

In ultrafast optics ultrashort pulses are routinely analyzed spectrally and temporally, but time-domain characterization tools are not easy to implement for single photons. Recently we have introduced a time-resolved single-photon measurement technique by use of femtosecond up-conversion [11]. In this Letter we utilize this single-photon time-domain characterization method to measure for the first time the joint temporal density (JTD) profile of a two-photon quantum state. In particular, we measured directly the temporal correlations of signal-idler arrival times of ultrafast-pumped SPDC under extended phase-matching conditions [5], showing clearly that the coincident-frequency entangled photons were time anticorrelated. Furthermore, by varying the SPDC pump spectrum, we were able to manipulate the temporal correlations of the signal and idler, and obtain a nearly unentangled (temporally) two-photon state. This new technique can be used in conjunction with frequency-domain methods to provide a more complete characterization of single and entangled photons.

To properly define JTD, we first express the two-photon state in time-domain variables $|\Psi\rangle = \iint d\tau_S d\tau_I \mathcal{A}(\tau_S, \tau_I)|\tau_S\rangle|\tau_I\rangle$, where $|\tau_j\rangle \equiv \hat{a}^\dagger(\tau_j)|0\rangle$ is the single-photon Fock state for $j = S, I$. The temporal correlations of the signal (S) and idler (I) are determined by the joint temporal amplitude, $\mathcal{A}(\tau_S, \tau_I)$, and the joint temporal density is given by $|\mathcal{A}(\tau_S, \tau_I)|^2$. Analogous to the frequency-domain methods, the JTD can be measured using narrow band *temporal* filtering and coincidence detection. Single-photon timing resolution of $\sim$100 fs is needed for typical ultrafast SPDC experiments. Current single-photon detectors with tens of picoseconds timing resolution are not suitable for this purpose. For the two-photon JTD measurement, we applied our recently developed time-resolved single-photon up-conversion technique with a temporal resolution of $\sim$150 fs [11]. In this method, an ultrafast up-converting pump pulse is used to time stamp the signal and idler arrival times, and their relative arrival times are mapped by varying the input delay lines independently and recording the coincidences between the two up-converted outputs. The coincidence statistics yields the temporal structure of the two-photon state. We note that two-photon second-harmonic coincidence measurements can also yield femtosecond-scale two-photon coherence time information [12].

Our experimental setup for ultrafast type-II phase-matched SPDC and subsequent JTD measurement with time-resolved up-conversion is shown in Fig. 1(a). Both SPDC and up-conversion were pumped synchronously with the same ultrafast source at 790 nm with a 6-nm bandwidth and 80 MHz repetition rate, thereby eliminating the pump timing jitter for the JTD measurement. We operated the 1-cm periodically poled $KTiOPO_4$ (PPKTP) SPDC crystal (46.1 μm grating period, 58 °C) under extended phase-matching conditions to generate a coincident-frequency entangled two-photon state [3,5]. By Fourier duality, this positive frequency correlation corresponded to anticorrelation in the time domain where the signal and idler photons with ~350-fs single-photon coherence times were symmetrically located about the center of a ~1.4-ps two-photon coherence time window, as measured by HOM interference [5]. The signal and idler photons were coupled into a polarization-maintaining (PM) single-mode fiber and separated at a fiber polarizing beam splitter. The signal and idler delay lines (55-cm PM fibers) were individually adjusted so that they arrived at the up-conversion crystal in the same time slot as the pump pulse. Fine-tuning of the relative timing was achieved with translation stages. We estimate that fiber-induced dispersive broadening would contribute to less than 10% of the single-photon coherence length.

We utilized the same time-resolved single-photon up-conversion setup as in Ref. [11], as shown in Fig. 1(b) and briefly described here. A 1-mm long periodically poled MgO-doped stoichiometric lithium tantalate (PPMgSLT) crystal with an 8.5 μm grating period was used for noncollinear type-0 phase-matched sum-frequency generation, up-converting the 1580-nm signal and idler to outputs at 526.7 nm. The noncollinear geometry was utilized to implement two independent up-converters with a single crystal. The signal and idler beams were each aligned at an angle of 2° relative to the pump beam, as shown in Fig. 1(b). We tilted the two input beams vertically to form a nonplanar geometry for the pump and the input signal and idler beams. The nonplanar focusing configuration allowed us to avoid the simultaneous detection of non-phase-matched down-converted photon pairs that were both generated and up-converted by the pump at the PPMgSLT crystal. Therefore, even with a finite background for singles, the coincidence profile shows negligible accidentals [11]. The up-converted outputs were filtered by dichroic mirrors and 10-nm bandpass interference filters centered at 530 nm, coupled into single-mode fibers and detected with fiber-coupled Si avalanche photodiodes. Singles and coincidence counts were recorded using a 1.8-ns coincidence window. We achieved an internal up-conversion efficiency of 25% limited by the available up-conversion pump power of 580 mW [11]. The up-conversion probability per pump pulse was actually lower because the pump pulse was much shorter than the effective pulse width of the signal and idler. The temporal resolution of ~150 fs was limited by the pump pulse width [11].

We measured the singles and coincidences by scanning the up-conversion pump pulse delay relative to the signal and idler arrival windows, and each data point was an average of 60 1-s measurements. The normalized histograms are plotted in Fig. 2 without background subtraction. For the optimal pump power ratio (~360 mW for down-conversion, ~580 mW for up-conversion) the maximum singles (coincidence) rate at the center of the distribution was ~5300/s (~17/s), including the background. The background level in singles counts were ~1900/s for the optimal pump power ratio, corresponding to a background

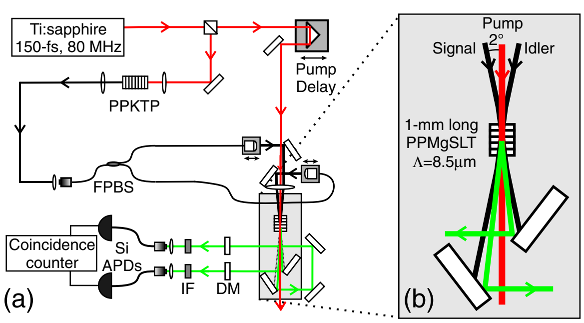

FIG. 1 (color online). (a) Synchronized up-conversion and down-conversion experiment driven by the same ultrafast pump. (b) Noncollinear phase-matching geometry for single-photon up-conversion. IF: interference filter; DM: dichroic mirror; FPBS: fiber polarizing beam splitter; APD: avalanche photodiode.

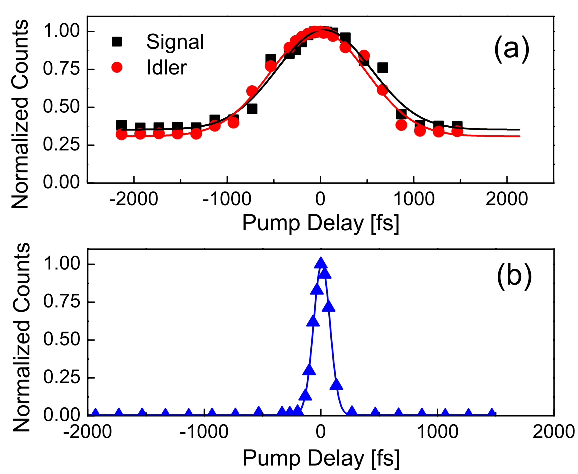

FIG. 2 (color online). Normalized singles (a) and coincidence (b) histograms by time-resolved up-conversion. The pump pulse was scanned through collocated signal and idler arrival windows. Solid lines are Gaussian fits to the data.

probability per pulse of $\sim 2.4 \times 10^{-5}$. The temporal width for singles distribution was $\sim$1.3 ps, consistent with our previously measured two-photon coherence time [5]. The coincidence profile of Fig. 2(b) shows a full width at half maximum (FWHM) of $\sim$165 fs that is much narrower than the singles histograms. In obtaining the coincidence curve, the up-conversion pump delay was scanned through the signal and idler arrival windows and, given that the signal and idler photons were anticorrelated in time, simultaneous up-conversion of the signal and idler photons could only occur near the time origin, thus producing the observed narrow coincidence peak.

To manipulate the joint temporal amplitude without affecting the up-conversion setup, we modified only the SPDC pump bandwidth by inserting a set of interference filters to yield a 3-dB bandwidth of 3.6 nm, 2.1 nm, or 1.1 nm before the PPKTP SPDC crystal. Figure 3 plots the normalized coincidence histograms with different SPDC pump bandwidths, clearly showing that as the SPDC pump bandwidth was reduced, the single-photon coherence time increased and consequently the coincidence peaks became wider.

The measurements in Fig. 3 are in good agreement with the theoretical coincidence profiles (dashed lines) which we calculate in the following way. First, we measured the various pump spectra with an optical spectrum analyzer. Second, we assume the standard sinc phase-matching function with a bandwidth given by our previous HOM measurement [5]. The product of the pump spectrum and the phase-matching function gives a theoretical joint spectral amplitude (JSA) [3] that assumes a zero spectral phase. The JTD is the modulus square of the Fourier transform of the JSA. Finally, a convolution of the JTD and the 150-fs up-conversion pump pulse produces the theoretical coincidence profiles of Fig. 3. A zero spectral phase implies that the theoretical curves assume transform limited two-photon states. The good agreement between data and the calculated values in Fig. 3 therefore suggests that dispersive broadening was negligible and that our assumption of a transform limited state is justified.

We utilized the up-conversion technique to measure the joint temporal density by independently varying the signal and idler time delays relative to the up-conversion pump pulse. The normalized coincidence data at different SPDC pump bandwidths are shown as surface and contour plots in Fig. 4(a)–4(d). The grid size in Fig. 4 was 2×2 ps with a 133 fs measurement step size, except for Fig. 4(d) (1.1-nm pump bandwidth) in which we used a 4×4 ps grid with a 266 fs step size. Each data point in Fig. 4 was an average of 60 1-s measurements. Figure 4 reveals a dramatic change in the JTD profile as a function of the SPDC pump bandwidth. With a 6 nm SPDC pump bandwidth, the JTD coincidence profile in Fig. 4(a) clearly exhibits time anticorrelation that is indicative of two-photon coincident-frequency entanglement [5]. At the smallest SPDC pump bandwidth of 1.1 nm, the JTD distribution in Fig. 4(d) is more symmetric, corresponding to reduced temporal (and spectral) correlation.

We can quantify the two-photon frequency entanglement as a function of the pump bandwidth based on the measured JTD distributions and by using Schmidt decomposition for continuous variables [13]. In this formalism, the joint temporal amplitude $\mathcal{A}(\tau_S, \tau_I)$ is expressed as a discrete sum of the temporal eigenmodes with eigenvalues λ_n, through which one can compute the entanglement entropy $S = -\sum_{k=0}^{n} \lambda_k \log_2 \lambda_k$ [13]. Figure 5 plots S computed from the experimental JTD distributions of Fig. 4, assuming that the joint state is transform limited. For comparison, we have also calculated the theoretical en-

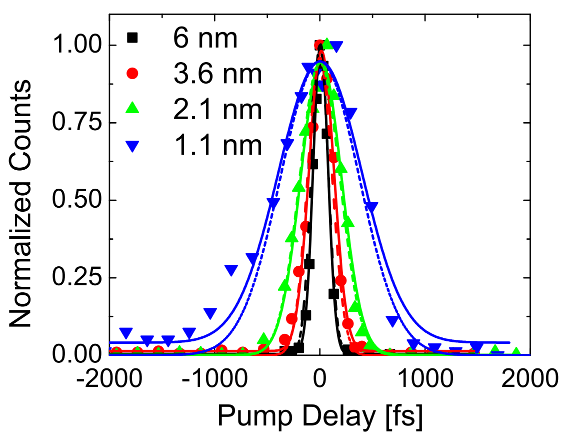

FIG. 3 (color online). Normalized coincidence histograms for various SPDC pump 3-dB bandwidths: (6-, 3.6-, 2.1-, and 1.1-nm). Solid lines are best fits to data. Theoretical coincidence profiles are plotted as dashed lines.

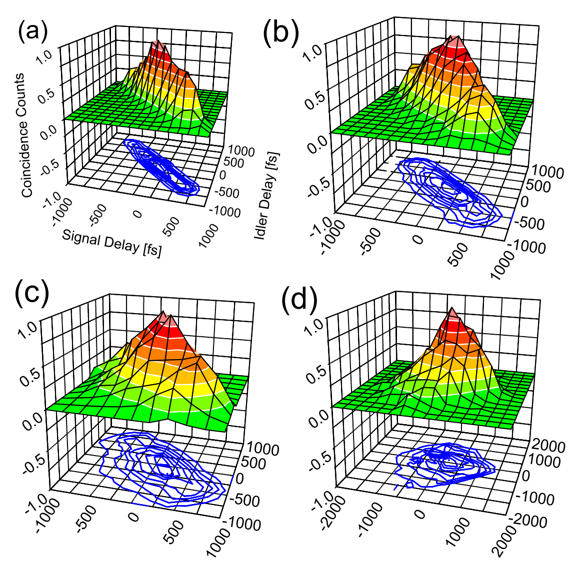

FIG. 4 (color online). Experimental joint temporal densities for various down-conversion pump 3-dB bandwidths: (a) 6 nm, (b) 3.6 nm, (c) 2.1 nm, (d) 1.1 nm.

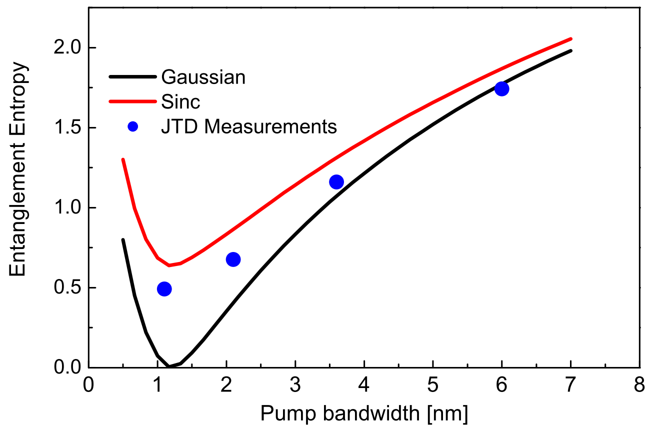

FIG. 5 (color online). Entanglement entropy values calculated from experimental JTD distributions for various SPDC pump bandwidths of Fig. 4. The theoretical entropy variations for Gaussian (black) and sinc-type (red) phase-matching functions are given in solid curves.

tropy curves as a function of the SPDC pump bandwidth for a Gaussian pump spectrum. Two curves are plotted in Fig. 5, one representing a Gaussian and the other a sinc phase-matching function. For a Gaussian phase-matching function, a fully factorizable two-photon state with $S = 0$ is predicted at a pump bandwidth of $\sim$1.2 nm. For the more realistic sinc phase-matching function, a highly but not completely factorizable two-photon state is achievable. Since the sinc-type spectral response corresponds to a boxcar shape in the time domain, it necessitates the inclusion of higher order Schmidt modes and hence increases the entanglement entropy.

Figure 5 shows a good qualitative agreement between the theoretical entropy curves and the entropy values obtained from the JTD distributions. The entanglement entropy corresponding to the experimental JTD profiles are lower than the theoretical curve for the sinc phase-matching function. This is reasonable if we take into account that the actual time-domain profile of the phase-matching function is smoother than a boxcar shape because of grating inhomogeneity, as confirmed by the singles histogram measurements of Fig. 2. Therefore, the experimental JTD distributions can be expressed with a smaller number of Schmidt modes, resulting in a lower entanglement entropy than that of the sinc-function case. For a 1.1-nm SPDC pump bandwidth with a nearly factorizable output, we compute the purity of the heralded single-photon state to be 0.88, where purity is defined as $p = \mathrm{Tr}(\hat{\rho}_S^2) = \sum_{n=0}^{\infty} \lambda_n^2$ [6,13]. This purity value compares well with that of the pure-state single photons generated from SPDC using a different spectral engineering method [7]. We believe that the purity can be further improved by finer control over the pump bandwidth and additional spectral filtering. In comparison, the output for the case of a 6-nm SPDC pump bandwidth yields a purity of 0.38 due to the high degree of coincident-frequency entanglement.

In conclusion, we have utilized a time-resolved single-photon measurement technique with subpicosecond resolution to measure for the first time the joint temporal density of a two-photon state. We applied the technique to verify anticorrelation in the arrival times of the coincident-frequency entangled signal and idler photons. Finally, the new tool allowed us to monitor the effect of varying the SPDC pump bandwidths, leading to the generation of a nearly factorizable two-photon state, which should be of interest to many quantum information processing applications. We believe that the JTD measurement technique is a powerful tool for engineering temporal and spectral correlations of ultrafast SPDC photons.

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