Joint Temporal Density Measurements for Two-Photon State Characterization

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We demonstrate a new 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 anti-correlation 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 in quantum information processing.

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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 (LOQC) [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, which can be a valuable resource for LOQC [3,7].

Characterizing the spectral correlations of a two-photon state can be done by measuring the joint spectral density (JSD) profile with tunable narrowband filtering of the signal and idler [3,7,8]. Hong-Ou-Mandel quantum interference [2] 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 in wavelength regions 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 upconversion [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 time correlations of signal-idler arrival times of ultrafast pumped SPDC under extended phase matching conditions [2], showing clearly that the coincident-frequency entangled photons were time anti-correlated. 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 |Ψ⟩ = ∫∫ dτSdτI A(τS, τI)|τS⟩|τI⟩, where the single-photon Fock state is defined as |τ⟩ = a†(τ)|0⟩, for j = S, I. The temporal correlations of the signal and idler are determined by the joint temporal amplitude, A(τS, τI), and we define the associated probability density, ⟨|A(τS, τI)|2⟩, as the joint temporal density. Analogous to the frequency-domain methods, the JTD can be measured by using narrowband temporal filtering and coincidence detection. For typical ultrafast SPDC experiments, timing resolution of ~100 fs is needed for measuring arrival times of single photons. 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 upconversion technique with a temporal resolution of \(\sim 150 \text{fs} \) \([11]\). An ultrafast upconverting pump pulse was used to time-stamp the signal and idler arrival times, and we mapped their relative arrival times by varying the input delay lines independently and recording the coincidences between the two upconversion channels. The coincidence statistics yielded the temporal structure of the two-photon state.

Our experimental setup for ultrafast type-II phase-matched SPDC and subsequent JTD measurement with time-resolved upconversion is shown in Fig. 1(a). Both SPDC and upconversion 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 PPKTP SPDC crystal under extended phase-matching conditions to generate a coincident-frequency entangled two-photon state \([3, 4]\). By Fourier duality, this positive frequency correlation corresponded to anti-correlation in the time domain where the signal and idler photons with \(\sim 350-\text{fs} \) single-photon coherence times were symmetrically located about the center of a \(\sim 1.4-\text{ps} \) two-photon coherence time window, as measured by HOM interference \([3]\). The signal and idler photons were coupled into a polarization-maintaining single-mode fiber and separated at a fiber polarizing beam splitter. The signal and idler delay lines were individually adjusted so that they arrive at the upconversion crystal in the same time slot as the pump pulse. Fine tuning of the relative timing can be achieved with translation stages.

We used the same setup as in Ref. \([11]\) for time-resolved single-photon upconversion, briefly described here. As sketched in Fig. 1(b), a 1-mm long periodically poled MgO-doped stoichiometric lithium tantalate (PPMgSLT) crystal with a 8.5 \(\mu\text{m} \) grating period was used for noncollinear type-0 phase-matched sum-frequency generation (1580 nm + 790 nm \(\rightarrow\) 526.7 nm). We used the noncollinear geometry to implement two independent upconverters with a single crystal. The single-photon beams were aligned parallel to the pump beam with \(\sim 3 \text{ mm}\) lateral and \(\sim 1.5 \text{ mm}\) vertical separation from the pump axis, and they were focused into the PPMgSLT crystal. The non-planar focusing configuration allowed us to avoid the simultaneous detection of the non-phase-matched parametric photon pairs that were both generated and upconverted by the pump at the PPMgSLT crystal. Therefore, even with a finite background for singles, the coincidence profile shows negligible accidentals \([11]\). The upconverted outputs were filtered by dichroic mirrors and 10-nm pass-band interference filters, coupled into single-mode fibers and detected with fiber-coupled Si APDs. We recorded the singles counts and also the coincidence counts between the two Si APDs within a 1.8 ns coincidence window.

We measured the singles and coincidences by scanning the upconversion pump pulse delay relative to the signal and idler arrival windows, and each data point was averaged for 60 seconds. The normalized histograms are plotted in Fig. 2 without any background subtraction. For the optimal pump power ratio (\(\sim 360 \text{ mW} \) for downconversion, \(\sim 580 \text{ mW} \) for upconversion) the maximum singles (coincidence) rate at the center of the distribution was \(\sim 5300/s \) \((\sim 17/s)\), including the background. The background level in singles counts were \(\sim 1900/s \) for the optimal pump power-ratio, corresponding to a background probability per pulse of \(\sim 2.4\times 10^{-5}\). The temporal width for singles distribution was \(\sim 1.3 \text{ ps}\), consistent with the two-photon coherence time of \(\sim 1.4 \text{ ps} \) \([3]\). Due to the time anti-correlated generation of signal and idler, the coincidence profile exhibited a \(\sim 165 \text{ fs}\) FWHM width, which is significantly narrower than the singles

![FIG. 1: (Color online) (a) Synchronized upconversion and downconversion experiment driven by the same ultrafast pump. (b) Noncollinear phase-matching geometry for single-photon upconversion. IF: interference filter; DM: dichroic mirror; FPBS: fiber polarizing beam splitter.](image)

![FIG. 2: (Color online) Normalized singles (a) and coincidence (b) histograms by time-resolved upconversion. The pump pulse was scanned through collocated signal and idler arrival windows. Solid lines are Gaussian fits to the data.](image)
histograms. As the upconversion pulse was scanned through the arrival windows of both photons, the only instance where the two upconverters could simultaneously detect photons was around the time origin. For an upconversion pump power of 580 mW, the internal conversion efficiency was estimated to be 25% \[1\]. However, the upconversion probability per pump pulse was actually lower because the pump pulse was much shorter than the effective pulse width of the signal and idler.

In order to manipulate the joint temporal amplitude without affecting the upconversion timing performance, we modified only the SPDC pump bandwidth by inserting a filter from a set of interference filters (3-dB bandwidths: 3.6 nm, 2.1 nm, and 1.1 nm) before the PP-KTP crystal. The measured normalized coincidence histograms for different SPDC pump bandwidths are plotted in Fig. 3. As the SPDC pump bandwidth was reduced, the single-photon coherence time increased and consequently the coincidence peaks became wider. In the same figure, we also show the theoretical predictions for the coincidence histograms that we calculate based on the joint temporal amplitude with a finite-duration upconversion pump pulse. The parameters for the calculation are the upconversion and downconversion pump bandwidths and the two-photon coherence bandwidth that we measured with the HOM interference \[4\]. We assume a flat spectral phase profile in our joint temporal amplitude calculation leading to predicted temporal coincidence profiles that suggest transform-limited two-photon states. The good agreement in Fig. 3 between data and theory indicates that the SPDC output photon pairs were indeed close to the transform limit. This observation is only possible with time-domain measurements because frequency-domain methods would be insensitive to dispersive broadening of the photons.

The time-resolved upconversion method enabled us to measure the joint temporal density by varying the signal and idler relative delays independently. We set the upconversion pump bandwidth to \(~6\) nm, and we made the JTD measurements using one of the four SPDC pump bandwidths. The coincidence counts were recorded over a two-dimensional time grid with 60-s averaging for each data point. For all SPDC pump bandwidths except 1.1 nm, the grid size for the time delays was set to \(2\) ps \(\times\) \(2\) ps (with a 133 fs step size). We increased the grid size to \(4\) ps \(\times\) \(4\) ps (266 fs step size) for the 1.1 nm pump bandwidth. The normalized coincidence data for all SPDC pump bandwidths are shown as surface plots over the two-dimensional time grids in Fig. 4(a)-(d). We see dramatic changes in the JTD profile with a change of the SPDC pump bandwidth. With a \(6\) nm SPDC pump bandwidth the JTD coincidence profile clearly exhibits time anti-correlation that is indicative of two-photon coincident-frequency entanglement \[4\]. With smaller pump bandwidths, the JTD distributions become more symmetric, which corresponds to reduced temporal and spectral correlations.

![FIG. 3: (Color online) Normalized coincidence histograms for various SPDC pump 3-dB bandwidths: (6 nm, 3.6 nm, 2.2 nm, and 1.1 nm). Theoretical coincidence profiles are plotted as dashed lines.](image)

![FIG. 4: (Color online) Experimental joint temporal densities for various downconversion pump 3-dB bandwidths: (a) 6 nm, (b) 3.6 nm, (c) 2.2 nm, (d) 1.1 nm.](image)

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 \[12\]. In this formalism, the joint temporal amplitude, \(A(\tau_s, \tau_i)\), is expressed as a discrete sum of the temporal eigenmodes with eigenvalues \(\lambda_k\), through which the entanglement entropy can be computed as \(S = -\sum_{n=1}^{\infty} \lambda_k \log_2 \lambda_k\) \[12\]. Figure 3 shows the computed entanglement entropy from the experimental JTD distributions in Fig. 4 assuming that the joint state is transform limited. For comparison, we have
also calculated the theoretical entropy curves as a function of the SPDC pump bandwidth, where the pump spectrum is assumed to be Gaussian. 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 is predicted with a pump bandwidth of \( \sim 1.2 \text{ nm} \), and yielding an entropy of zero. For the more realistic sinc function for the phase-matching, 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 theoretical of a sinc function. For a 1.1-nm SPDC pump bandwidth, which yields an output that is nearly factorizable, we have computed the purity of the heralded single-photon state as \( \rho = \frac{1}{2} + \frac{1}{2} S \sum_{n=0}^{\infty} \lambda_n^2 \approx 0.38 \), which is a consequence of the high degree of coincident-frequency entanglement.

In conclusion, we have developed a time-domain measurement technique for single photons with sub-picosecond resolution that we used to measure the two-photon joint temporal density for the first time. We applied the technique to verify anti-correlation in the arrival times of the signal and idler photons that were coincident-frequency entangled. 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. Such a characterization technique would complement the frequency-domain counterparts to quantify and manipulate multi-photon entanglement for quantum information processing applications.

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.

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