Ultrafast quantum interferometry with energy-time entangled photons

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Many quantum advantages in metrology and communication arise from interferometric phenomena. Such phenomena can occur on ultrafast time scales, particularly when energy-time entangled photons are employed. These have been relatively unexplored as their observation necessitates time resolution much shorter than conventional photon counters. Integrating nonlinear optical gating with conventional photon counters can overcome this limitation and enable subpicosecond time resolution.

In this work, we temporally resolve two-photon interference with subpicosecond timing resolution. The detectors are implemented by optically gating the photons in a nonlinear medium via noncollinear sum-frequency generation (SFG) with a short gate pulse. Using this technique and single-photon spectrometers, we measure both the joint temporal and joint spectral features of a spatially separated two-photon state at the output of a Franson interferometer.

We produce energy-time entangled photon pairs with parametric downconversion pumped by a broadband laser pulse. The photons are produced with strong anti-correlations between the signal, ωs, and idler, ωi, frequencies leading to a narrow joint uncertainty Δ(ωs + ωi) set by the bandwidth of the pump in broadly phasematched materials. For a two-photon state with no spectral phase, the photon pairs will also exhibit strong correlations between the time of arrival of the signal, ts, and the idler, ti, leading to smaller joint uncertainty than

Interferometry based on entangled quantum states is essential for enhanced metrology and quantum communication. Quantum correlations can enable interferometric measurements with improved sensitivity and resolution, and quantum advantages have been found for interferometric applications involving optical coherence tomography; precise measurements of optical properties; and even the detection of gravitational waves. In laser physics, the development of ultrafast light sources has led to innovations in atomic spectroscopy, time-resolved measurements for quantum chemistry, nonlinear optics, x-ray sources, with applications in health sciences and industrial machining.

For quantum light, energy-time entangled photons can also be produced with temporal features on ultrafast time scales and the wide availability of pulsed lasers has made this regime accessible for quantum state engineering. However, quantum interferometry with these states is challenging as the interference time scales are below the resolution of standard photon detectors. To overcome detector limitations, optical techniques have been developed to directly observe energy-time entangled quantum states on ultrafast time scales by building effective fast photon counters using ultrafast optical gating in conjunction with standard photon counters. Extending the measurement of quantum interference to subpicosecond time scales will be essential for developing new applications with ultrafast energy-time states of light.

An important class of interferometers that has been used to observe quantum interference effects with energy-time entangled photons was developed by Franson in 1989. Photon pairs are sent through two unbalanced interferometers creating interference in the coincidence rate but not in the single-photon detection rates. High-visibility interference was observed in such an interferometer using spontaneous parametric downconversion (SPDC). Franson interferometers with energy-time entangled states have since become important for applications in long-distance quantum key distribution, measuring entanglement in high-dimensional and multiphoton states, scaling quantum information tasks to larger dimensions, and improving molecular spectroscopy. When both the single-photon and two-photon coherence times are ultrafast, however, observing quantum interference effects with a Franson interferometer requires new techniques to overcome detector limitations and the original interferometer concept can be adapted to provide delays on shorter time scales.
their individual widths in time, $\Delta(t_s - t_i) < \Delta t_{s,i}$ [16]. A Franson interferometer, shown schematically in Fig. 1(a), separates the photons on each side into a short and long path, with a time delay $\tau$, resulting in four possible combinations of paths. The single-photon detection rates, which vary with the phase in each arm, $\phi_{s,i}$, have a coherence time inversely proportional to the single-photon spectral bandwidth, $\tau_{c,s,i}^{(1)} = 1/\Delta \omega_{s,i}$, whereas the coincidence rate, which varies with $\phi_s + \phi_i$, has a two-photon coherence time inversely proportional to the two-photon spectral bandwidth, $\tau_{c,s,i}^{(2)} = 1/\Delta(\omega_s + \omega_i)$ (see supplemental material). When $\tau$ is set to be much larger than the single-photon coherence time but less than the two-photon coherence time, $\tau_{c,s,i}^{(1)} \ll \tau < \tau_{c,s,i}^{(2)}$, interference in the coincidence rate can be observed without any present in the single detection rates.

The interference in the coincidences results from the indistinguishability between the cases where the two photons both take the short path in the interferometer and where both take the long path. Meanwhile, the cases where they take opposite paths, labeled short-long and long-short, do not exhibit interference, thus limiting the visibility to 50% without temporal resolution. This, however, is the same maximum visibility that can be obtained in coincidence measurements with classically correlated light when zero visibility is observed in the single-photon rates [32]. To observe higher visibility interference with energy-time entangled photons, sufficient time resolution is needed to resolve the arrival times of the early and late photons. This condition is typically met using continuous-wave-pumped downconversion sources whose two-photon coherence times are much longer. They can therefore support interferometer delays in the range of 10 cm to 1 m [33–34], which can be implemented in free space or fiber, such that the time difference $\tau$ between early and late photons (30 $\mu$s to 3 ns) remains much larger than standard detector resolution.

We construct the ultrafast Franson interferometer using birefringent crystals where the long and short paths arise due to the different refractive indices, and hence different optical path lengths, for horizontally and vertically polarized light [37], as seen in Fig. 1(b). Two millimeters of $\alpha$-BBO creates an interferometer with relative delays below one picosecond and does not require

![Franson interferometer concept diagram](image)

**FIG. 1.** Franson interferometer concept diagram. (a) Nonlocal interference can be seen by sending each photon of a two-photon energy-time entangled pair through an unbalanced interferometer. Each photon is split into early and late time bins and recombined with a phase applied to one bin. (b) In each arm of the Franson interferometer, the delays and phases are implemented through birefringent material and wave-plates, creating a path difference on subpicosecond time scales between the short and long paths. A birefringent crystal ($\alpha$-BBO) splits a horizontally polarized photon into a diagonal and a delayed anti-diagonal mode. A quarter-wave plate (QWP) converts diagonal and anti-diagonal to left- and right-circularly polarized light. A half-wave plate (HWP) introduces a phase between the circularly polarized photons. Both polarizations are then projected into the horizontal state with a polarizing beam splitter (PBS).

![Experimental setup](image)

**FIG. 2.** Experimental setup. (a) A Ti:sapphire laser pulse (775 nm, 3.8W average power, 0.120 ps (s.d.) pulse-width), is frequency doubled in 2 mm of $\beta$-bismuth borate (BiBO). After spectral filtering with a 0.2 nm FHWM bandpass filter, the second harmonic (387.6 nm, 300 mW average power, approximately 0.940 ps (s.d.) coherence time) pumps a 5 mm BiBO crystal for type-I spontaneous parametric downconversion (SPDC) generating frequency entangled photons centered at 730 nm and 827 nm. The photons are separated by a dichroic mirror and their bandwidth is controlled using tunable edge filters. Each photon passes through an unbalanced interferometer consisting of $\alpha$-BBO, QWP, HWP, and PBS. We use 2.00 mm and 2.25 mm of $\alpha$-BBO to create a difference between the short and long paths of $\tau_s = 0.820$ ps and $\tau_i = 0.910$ ps on the signal and idler side, respectively. The output of the Franson interferometer is coupled into single-mode fibers. (b) Spectral measurements are made with single-photon spectrometers. (c) Temporal measurements are performed using ultrafast gating with a strong laser pulse. A pair of grating compressors compensates for the dispersion introduced by the fibers.
FIG. 3. The joint spectral intensity and joint temporal intensity of the two-photon state shown (a,d) before and (b,c,e,f) after the Franson interferometer. After the interferometer, different fringe patterns are observed in the joint spectrum for (b) constructive and (c) destructive interference. The interferometer shifts the temporal profile in (d) creating four different combinations of paths: short-short, short-long, long-short, and long-long. We observe (e) constructive and (f) destructive interference in the central peak between the cases where the photons both take the short path and both take the long path. These correspond to two-photon states where the signal and idler phases sum to (b,e) $\phi_i + \phi_s = 0$, and (c,f) $\phi_i + \phi_s = \pi$.

any active phase stabilization. The experimental setup is shown in Fig. 2. Signal-idler photon pairs are produced using SPDC with center wavelengths of 730 nm and 827 nm, respectively. A pair of tunable edge filters in the source control the single-photon spectral bandwidths by making effective bandpass filters of 3.0 nm (s.d.) and 3.5 nm, for the signal and idler respectively. The photon pairs are coupled into fiber, allowing for direct detection, spectral, or temporal measurements in coincidence, with or without the Franson interferometer. Spectral measurements are performed using two grating-based single-photon monochromators with a resolution of approximately 0.1 nm, while temporal measurements are implemented by optically gating the single-photons using SFG with femtosecond laser pulses which have an intensity pulse width of 0.120 ps (s.d.) [16].

The joint spectral intensity and joint temporal intensity of the state before the Franson interferometer were measured and the data is shown in Figs. 3(a) and 3(d), respectively. We observe strong anti-correlations between the photon frequencies and strong positive correlations between their arrival times. Measurements of the spectral widths in these plots allow us to estimate the one- and two-photon coherence times. To account for the finite resolution of the spectrometers and temporal gates, Gaussian fits to the measured widths are deconvolved assuming Gaussian response functions. The deconvolved frequency marginals (see supplemental material) are found to be $\Delta \omega_s = 10.65$ ps$^{-1}$ and $\Delta \omega_i = 9.57$ ps$^{-1}$, from which we estimate single-photon coherence times of $\tau^{(1)}_c = 0.094$ ps for the signal and $\tau^{(1)}_i = 0.105$ ps for the idler respectively. Gaussian fits to histograms of the spectral semi-minor and semi-major axes yield deconvolved two-photon spectral bandwidths of $\Delta (\omega_s + \omega_i) = 1.531$ ps$^{-1}$ and $\Delta (\omega_s - \omega_i) = 17.81$ ps$^{-1}$. From the former, we estimate a two-photon coherence time of $\tau^{(2)} = 0.653$ ps. The temporal measurements yield deconvolved temporal marginal widths of $\Delta t_s = 0.455$ ps and $\Delta t_i = 0.488$ ps and deconvolved temporal widths of the semi-minor and semi-major axes of $\Delta (t_s + t_i) = 0.895$ ps and $\Delta (t_s - t_i) = 0.091$ ps.

The joint spectral intensity and joint temporal intensity of the state after the Franson interferometer are shown in Figs. 3(b, c, e, f) for two different combinations of phase settings which provide the highest contrast between the constructive and destructive interference in the central peak of the temporal plots. In Fig. 3(b), we observe a joint spectral intensity similar to the one found in Fig. 3(a) but with a periodic amplitude modulation. The joint spectral intensity in Fig. 3(c) is also modulated
The coincidence rate exhibits fringes with \( (85.4 \pm 0.4)\% \) visibility, while the singles detection events show no apparent oscillations.

To maximize the visibility of the Franson interference, we found that the interferometer delays need to shift the joint-temporal intensity in Fig. 3(d) along its semi-major axis, such that a maximum overlap is obtained between the cases where both photons take the long path and where both photons take the short path. This can be achieved by matching the ratio of the applied interferometer delays \( \tau \) to the ratio of the marginal temporal widths \( \Delta t \), such that \( \tau_s / \tau = \Delta t_s / \Delta t_i \). The measured ratio was \( \Delta t_i / \Delta t_s = 1.07 \), differing from unity due to the particular phase-matching conditions which can change the angle of the joint spectral amplitude function \( R_{\phi} \).

We found that using different lengths of \( \alpha \)-BBO crystals, 2.00 mm and 2.25 mm, created the appropriate temporal separations of approximately \( \tau_s = 0.820 \text{ ps} \) and \( \tau_i = 0.910 \text{ ps} \), for the signal and idler, respectively, in order to approach this ratio and satisfy the conditions for two-photon interference. We repeated the measurement when both crystal lengths were chosen to be 2.00 mm and observed a reduction of 10\% in the visibility.

The measured detector counts for each phase setting on the signal and idler sides can be viewed as one binary outcome (+1) of a projective measurement, where the corresponding outcome (−1) is obtained by a \( \pi \) phase shift. Thus, we can look for a violation of the CHSH inequality from 16 combinations of signal-idler phases, 4 outcomes for each of the 4 joint projective measurements in the inequality \( 24 \) (see supplemental material). We count for 200 seconds for each outcome, and obtain from these counts a CHSH-Bell parameter of \( 2.42 \pm 0.02 \), a violation of the local-hidden variable bound of 2 by 21 standard deviations \( 29 \). This is a consequence of the entanglement in our system and shows the high quality of the interference and the general performance of our measurement device.

The visibility of the Franson interference and Bell violation could be further improved by reducing the second-harmonic generation (SHG) background from the laser in the optical gating. From the measured upconversion rates after the source, we obtain a coincidence rate of about 44 Hz from which about 0.8 Hz can be attributed to the SHG background of the laser. This corresponds to a signal-to-noise ratio (SNR) of 54. After the Franson interferometer, the measured coincidence rate at the peak is reduced by a factor of 4 but the SHG background remains the same, giving a SNR of 13.5. This translates to a reduction in visibility of 13\%, which accounts for most of the observed visibility loss. The SHG background source could be reduced by utilizing a type-II process which would allow for additional polarization filtering.

We have experimentally observed two-photon quantum interference on ultrafast time scales using a stable and compact Franson interferometer. The optical gating detection mechanism enables the direct measurement of the joint temporal intensity as well as the observation of quantum interference phenomena and the violation of a

![FIG. 4. Two-photon interference fringes. Franson interference between the upconverted signal and idler pair is measured by varying the signal and idler phases while setting the idler gate delay and signal gate delay halfway between the short and long paths of each side of the Franson interferometer. (a) We observe high-visibility interference with fringe oscillations along the diagonal which depend on the sum of the two phases \( \phi_s + \phi_i \). (b) Weighted average of the coincidences and weighted average of the singles for the signal and idler pair as viewed as a function of their phase sum. Interference fringes display oscillations of \( (85.4 \pm 0.4)\% \) visibility, while the singles detection events show no apparent oscillations.](image-url)
CHSH-Bell inequality in a previously inaccessible regime. In addition to interferometry, access to both spectral and temporal features will provide new tools for creating and characterizing two-photon states and will be essential for new applications in quantum state engineering, such as shaping ultrafast entangled photon pulses.

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SUPPLEMENTAL MATERIAL

The supplemental material is organized as follows. We first present additional fit parameters for the two-dimensional joint spectral intensity and joint temporal intensity in Figs. 3(a) and 3(d), respectively. We next describe the experimental implementation of the unbalanced interferometers in Fig. 1 and Fig. 2, and which were used to evaluate the CHSH inequality. We then consider the effect of finite correlations on the single-photon and two-photon detection rates at the output of the Franson interferometer.

Additional Experimental Details

Photons from the source were detected at a rate of 626,000 coincidence counts per second with $3.6 \times 10^6$ and $3.3 \times 10^6$ single-detection events per second for the signal and idler, respectively. The heralded second-order coherence of the source, measured with a Hanbury Brown-Twiss interferometer, was $g^{(2)}(0) = 0.391 \pm 0.004$ for the signal and $g^{(2)}(0) = 0.395 \pm 0.006$ for the idler. In general, double-pair emission will lead to a broad background in the joint spectrum and joint temporal intensity, however, due to the tight temporal filtering on both sides, we estimate that double pairs contribute to less than 1% of the measured up-converted signal. After the up-conversion on each side (without the Franson interferometer), approximately 44 coincidence counts (12,000 up-converted signal singles and 21,000 up-converted idler singles per second) per second were measured at the peak, from which about 0.8 coincidence counts (400 signal and 2,500 idler singles) per second were background from the second harmonic of the gate pulse.

We present the fit parameters for the measured widths of the joint spectral intensity in Fig. 3(a) and the joint temporal intensity in Fig. 3(d) of the main text. The marginal widths are obtained by fitting the marginals of Figs. 3(a) and 3(d) to a one-dimensional Gaussian, while the heralded widths are obtained taking the average of several slices of the data when the frequency or time of one photon is fixed. A visual representation of the marginal and heralded widths is presented in Fig. 5. The statistical correlation, $\rho$, is obtained by finding the value that best fits a two-dimensional Gaussian with the measured marginals. The fit parameters are deconvolved assuming a Gaussian response function (see supplemental material of Ref. [16]), and these values are presented in parentheses alongside the values obtained from the raw measurements in Table I.

| Property (Deconvolved) | Joint-spectrum | Joint-temporal intensity |
|-----------------------|----------------|-------------------------|
|                       | (Deconvolved)  |                          |
| Signal                |                |                          |
| Center Frequency ($\omega$) | 2584.6 $\pm$ 0.4 ps$^{-1}$ | -                        |
| Marginal width        | 10.65 $\pm$ 0.04 ps$^{-1}$ | 0.471 $\pm$ 0.004 ps    |
|                       | (10.63 $\pm$ 0.04 ps$^{-1}$) | (0.455 $\pm$ 0.004 ps) |
| Heralded width        | 1.25 $\pm$ 0.04 ps$^{-1}$ | 0.171 $\pm$ 0.009 ps    |
|                       | (1.13 $\pm$ 0.05 ps$^{-1}$) | (0.059 $\pm$ 0.022 ps) |
| Idler                 |                |                          |
| Center Frequency ($\omega$) | 2276.7 $\pm$ 0.4 ps$^{-1}$ | -                        |
| Marginal width        | 9.57 $\pm$ 0.04 ps$^{-1}$ | 0.502 $\pm$ 0.005 ps    |
|                       | (9.56 $\pm$ 0.04 ps$^{-1}$) | (0.488 $\pm$ 0.005 ps) |
| Heralded width        | 1.13 $\pm$ 0.02 ps$^{-1}$ | 0.183 $\pm$ 0.010 ps    |
|                       | (1.02 $\pm$ 0.02 ps$^{-1}$) | (0.063 $\pm$ 0.023 ps) |
| Statistical Correlation | $-0.9929 \pm 0.0001$ | 0.920 $\pm$ 0.003      |
|                       | ($-0.9942 \pm 0.0001$) | (0.979 $\pm$ 0.004)   |

TABLE I. Fit parameters for the joint spectral intensity and the joint temporal intensity as seen in Fig. 3(a) and 3(d) of the main text. All measured values are standard deviations and values in parentheses are deconvolved from a Gaussian response function.
The unbalanced interferometers for ultrafast photons

The experimental implementation of the Franson interferometer presented in Fig. 1 was chosen to provide a stable and compact method of creating time bin states with subpicosecond temporal separations. In this section, we analyze the transformations applied to the polarization state of the photon by the unbalanced interferometer in Fig. 1(b), composed of a birefringent crystal, wave plates, and a polarizing beam-splitter (PBS).

We denote the eigenstates of the Pauli operators $\sigma_z$ as $|H\rangle$ and $|V\rangle$, representing the horizontal and vertical polarization states of light. After downconversion, the polarization state of each photon is vertical, $|\psi\rangle_{pol} = |V\rangle$. The $\alpha$-barium borate ($\alpha$-BBO) birefringent crystals at 45 degrees separate the photons on each side into early, $|e\rangle$, and late, $|l\rangle$, time bins with a temporal separation of $\tau_e = 0.820$ ps and $\tau_l = 0.910$ ps, for the signal and idler, respectively. The two time bins have orthogonal polarizations, which we denote as diagonal, $|D\rangle = \frac{1}{\sqrt{2}} (|H\rangle + |V\rangle)$ and anti-diagonal, $|A\rangle = \frac{1}{\sqrt{2}} (|H\rangle - |V\rangle)$. As a result, the polarization state is transformed to, $|\psi\rangle_{pol} \rightarrow \frac{1}{\sqrt{2}} (|D\rangle |e\rangle + |A\rangle |l\rangle)$. The phase difference $\phi$ between the two time bins can be controlled by manipulating the polarization of the two modes after the $\alpha$-BBO crystals with two wave plates and a PBS. A quarter-wave plate (QWP) first converts the two orthogonal polarization modes into left-circular, $|L\rangle = \frac{1}{\sqrt{2}} (|H\rangle - i|V\rangle)$, and right-circular polarizations, $|R\rangle = \frac{1}{\sqrt{2}} (|H\rangle + i|V\rangle)$, resulting in the state $\frac{1}{\sqrt{2}} (|L\rangle |e\rangle + |R\rangle |l\rangle)$. A half-wave plate (HWP) at an angle $\theta$, described by the unitary operator,

$$U_{\text{HWP}}(\theta) = i \begin{pmatrix} \cos 2\theta & \sin 2\theta \\ \sin 2\theta & -\cos 2\theta \end{pmatrix},$$

FIG. 5. Spectral bandwidths and temporal widths for a frequency anti-correlated two-photon SPDC state. (a) The single-photon spectral bandwidth, $\Delta \omega$, is given by the marginal distribution obtained by projecting the joint spectral intensity onto either the signal or idler axes. The single-photon coherence time, the time scale over which interference in the single-photon rates can occur, is related to the inverse of the single-photon spectral bandwidth, $\tau^{(1)} = 1/\Delta \omega$. (b) The heralded spectral bandwidths, $\Delta \omega_h$, are the spectral bandwidths of the signal or idler photon when the frequency of the other is fixed. (c) The two-photon spectral bandwidths for the semi-minor, $\Delta (\omega_s + \omega_i)$, and semi-major axes, $\Delta (\omega_s - \omega_i)$, are obtained by projecting the joint spectral intensity along the corresponding diagonal axes $\omega_s \pm \omega_i$. The two-photon coherence time, the time scale over which interference in the coincidences can occur, is related to the inverse of the two-photon spectral bandwidth, $\tau^{(2)} = 1/\Delta (\omega_s + \omega_i)$. (d-f) The marginal temporal widths, $\Delta t$, the heralded temporal widths, $\Delta t_h$, as well as the two-photon temporal widths, $\Delta (t_s \pm t_i)$ are obtained from the joint temporal intensity in the same way as their spectral analogues.
next applies the following transformations on the left- and right-circular polarizations of light,

\[ U_{\text{HWP}}(\theta) | R \rangle = i e^{i \theta} | L \rangle \]
\[ U_{\text{HWP}}(\theta) | L \rangle = i e^{-i \theta} | R \rangle, \]
thus modifying the state to \( \frac{1}{\sqrt{2}} i \left( e^{i \theta} | R \rangle | e \rangle + e^{-i \theta} | L \rangle | l \rangle \). The PBS then erases the polarization information by projecting both circular polarizations into the horizontal mode \( | H \rangle \), transforming the state to \( \frac{1}{i} | H \rangle \left( e^{-i \theta} | e \rangle + e^{i \theta} | l \rangle \right) \). As a result of these transformations, the photon at the output of the unbalanced interferometer is in a time bin state with a phase difference between the early and late bins that can be set by the angle of the HWP through the parameterization \( \phi = 4 \theta \).

**Bell inequality using Franson interferometry**

The measured detector counts for each phase setting \( \phi \) in the unbalanced interferometers can be viewed as one binary outcome of a projective measurement, which we assign the value \((+1)\). The corresponding outcome \((-1)\) could be obtained by placing a second detector to measure the photon events at the second output port of the unbalanced interferometer, however, here the second outcome \((-1)\) is instead obtained by measuring the photon events from the same detector but with an additional \( \pi \) phase shift introduced in the interferometer using the HWPs in Fig. 2. Given measurement outcomes \( \pm 1 \) for two measurement choices labeled \( a, a' \) for the signal and \( b, b' \) for the idler, we measure the coincidence rates for the four outcomes of each joint projective measurement, denoted \( R_{i,j}(a,b), (i,j = \pm 1) \), and evaluate the correlation coefficient [24],

\[ E(a, b) = \frac{R_{++}(a,b) + R_{--}(a,b) - R_{+-}(a,b) - R_{-+}(a,b)}{R_{++}(a,b) + R_{--}(a,b) + R_{+-}(a,b) + R_{-+}(a,b)} \]

Assuming a local-hidden variable model, the CHSH inequality [39] provides an upper limit to the combination of four correlation coefficients, which can be written as,

\[ S = |E(a, b) + E(a, b') + E(a', b) - E(a', b')| \leq 2. \]

In Table II we provide a table of raw coincidence counts for a particular combination of two projective measurements in the \( x - z \) plane of the Bloch sphere on both the signal and idler sides. From these counts, a CHSH Bell-parameter of \( S = 2.42 \pm 0.02 \) is obtained, thus violating the inequality by 21 standard deviations.

**Franson interferometry with finite correlations**

In this section, we first calculate the overall coincident and single-photon detection rates of an energy-time entangled two-photon state after the Franson interferometer. We will show that this leads to two distinct time scales of interference for the single-photon detection rates and the coincidence detection rate. We then describe the need for
temporal selection to improve the visibility of the interference in the coincidence rate after the Franson interferometer, and describe the effect of spectral or temporal selection by calculating the joint spectrum and joint temporal intensity for two-photon state after the Franson interferometer. Finally, we discuss the parameters for optimizing the visibility of the two-photon interference.

Consider the two-mode state with signal \( \omega_s \) and idler \( \omega_i \) frequency modes,

\[
|\psi\rangle = \int d\omega_s d\omega_i F(\omega_s, \omega_i) a_{\omega_s}^\dagger a_{\omega_i}^\dagger |0\rangle .
\] (6)

At the source, the joint spectral amplitude \( F(\omega_s, \omega_i) \) of a pure two-mode state with no spectral phase can be described in Gaussian form as,

\[
F_{\text{source}}(\omega_s, \omega_i) = \frac{1}{\sqrt{2\pi \sigma_{\omega_s} \sigma_{\omega_i} (1 - \rho_s^2)^{1/4}}} \exp \left( -\frac{1}{2 (1 - \rho_s^2)} \left[ \frac{(\omega_s - \omega_{s0})^2}{2\sigma_{\omega_s}^2} + \frac{(\omega_i - \omega_{i0})^2}{2\sigma_{\omega_i}^2} - \frac{\rho_s (\omega_s - \omega_{s0})(\omega_i - \omega_{i0})}{\sigma_{\omega_s} \sigma_{\omega_i}} \right] \right),
\] (7)

where \( \sigma_{\omega_s} \) and \( \sigma_{\omega_i} \) are the marginal bandwidths of the signal and idler, respectively, and where the correlation parameter \( \rho_s = \Delta(\omega_s \omega_i) / \Delta \omega_s \Delta \omega_i \) describes the statistical correlations between the frequency of the signal and idler modes, and can be related to the purity of the partial trace, \( P = \sqrt{1 - \rho_s^2} \). For frequency anti-correlated photons, as shown in Fig. 3a), the frequency correlations are negative and \( \rho_s < 0 \).

The Franson interferometer introduces delays \( \tau_s \) and \( \tau_i \) between the short and long arms of each unbalanced interferometer with a phase \( \phi_s \) and \( \phi_i \) on the signal and idler sides, respectively. The joint spectral amplitude after the Franson interferometer takes the form,

\[
F_{\text{franson}}(\omega_s, \omega_i) = F_{\text{source}}(\omega_s, \omega_i) \times \frac{1}{4} \left( 1 + e^{i(\omega_s \tau_s + \phi_s)} \right) \left( 1 + e^{i(\omega_i \tau_i + \phi_i)} \right).
\] (8)

The overall coincidence rate directly after the interferometer is,

\[
C(\phi_s, \phi_i) = \int_{-\infty}^{\infty} d\omega_s d\omega_i |F_{\text{franson}}(\omega_s, \omega_i)|^2
\]

\[
\propto 1 + \exp \left( \frac{-\tau_s^2 \sigma_{\omega_s}^2}{2} \cos (\omega_{s0} \tau_s - \phi_s) \right) + \exp \left( \frac{-\tau_i^2 \sigma_{\omega_i}^2}{2} \cos (\omega_{i0} \tau_i - \phi_i) \right) + \frac{1}{2} \exp \left( -\frac{1}{2}(\sigma_s^2 \phi_s^2 - (1 + \rho_s) \sigma_s \sigma_i \tau_s \tau_i) \right) \cos \left( (\omega_{s0} \tau_s - \phi_s) + (\omega_{i0} \tau_i - \phi_i) \right)
\]

\[
+ \frac{1}{2} \exp \left( -\frac{1}{2}(\sigma_s^2 \phi_s^2 - (1 - \rho_s) \sigma_s \sigma_i \tau_s \tau_i) \right) \cos \left( (\omega_{s0} \tau_s - \phi_s) - (\omega_{i0} \tau_i - \phi_i) \right) .
\] (9)

For frequency anti-correlated photons, \( \rho \to -1 \), we expect interference which depends on the phase sum \( \phi_s + \phi_i \), whereas for frequency correlated photons \( \rho \to 1 \), the interference depends on the phase difference \( \phi_s - \phi_i \). Considering the idealized case of frequency anti-correlations (\( \rho \to -1 \)), assuming the signal and idler photon bandwidths \( \sigma_s \) are the same, and the interferometer delays \( \tau \) are equal, Eq. \ref{eq:9} simplifies to,

\[
C(\phi_s, \phi_i) \propto 1 + \exp \left( -\frac{\tau^2 \sigma_{\omega_s}^2}{2} \cos (\omega_{s0} \tau - \phi_s) \right) + \exp \left( \frac{\tau^2 \sigma_{\omega_i}^2}{2} \cos (\omega_{i0} \tau - \phi_i) \right) + \frac{1}{2} \exp \left( -(1 + \rho_s) \sigma_s^2 \tau^2 \right) \cos \left( (\omega_{s0} \tau - \phi_s) + (\omega_{i0} \tau - \phi_i) \right) .
\] (10)

On the other hand, single-photon detection events have interference fringes described by

\[
S(\phi_j) \propto 1 + \exp \left( -\frac{1}{2} \sigma_{\omega_j}^2 \tau_j^2 \right) \cos (\omega_{j0} \tau_j - \phi_j) .
\] (11)

where \( j \in \{s, i\} \). Comparing Eq. \ref{eq:10} and Eq. \ref{eq:11}, we find there are two time scales for interference for the two-photon state from downconversion. The single-photon interference in Eq. \ref{eq:11} varies with \( \phi_j \) and has a coherence time that depends on the inverse bandwidth of the photons, \( \tau_j^{(1)} = 1/\sigma_s = 1/\Delta \omega \), whereas the two-photon interference in Eq. \ref{eq:10} varies with the sum \( \phi_s + \phi_i \) and has a coherence time that depends on the two-photon spectral...
bandwidth, \( \tau^{(2)}_c = 1/(\sqrt{2}\sqrt{1 + \rho^2 \omega^2}) = 1/\Delta(\omega_s + \omega_l) \). The Franson interferometer can thus be used to separate these two time scales by setting the delay \( 1/\Delta \omega \leq \tau \leq 1/\Delta(\omega_s + \omega_l) \) between the single-photon and two-photon coherence times. Thus, with the appropriately chosen delay settings, we find the singles detection rates are constant whereas the coincident detection rate has oscillating fringes which depends on \( \phi_s + \phi_l \) with an interference visibility of \( V = \frac{1}{2} \exp\left(-\frac{1}{2} \Delta(\omega_s + \omega_l)^2 \tau^2\right) \). The visibility \( V \leq \frac{1}{2} \) without temporal selection is limited by the non-interfering background contributions from the short-long and long-short paths of the interferometer. In order to improve the measured visibility, these non-interfering background terms must be temporally filtered.

**Franson interferometry with spectral or temporal selection**

We now discuss the spectral and temporal features of the downconverted state after the Franson interferometer. This is achieved by calculating the joint spectrum and joint temporal intensity. The joint spectrum is obtained from the modulus squared of Eq.\( \text{[8]} \)

\[
|F_{\text{franson}}(\omega_s, \omega_l)|^2 = |F_{\text{source}}(\omega_s, \omega_l)|^2 \cos \left( \frac{\omega_0 \tau_s + \phi_s}{2} \right)^2 \cos \left( \frac{\omega_0 \tau_l + \phi_l}{2} \right)^2. \tag{12}
\]

It consists of the original source spectrum \( |F_{\text{source}}(\omega_s, \omega_l)|^2 \), which is intensity modulated. When \( \phi_l + \phi_s = 0 \), the oscillations for the anti-correlated frequencies remain in phase, as in Fig.\( \text{[3]} \)b) of the main paper, whereas when \( \phi_s + \phi_l = \pi \), they will be out of phase, as in Fig.\( \text{[3]} \)c).

The joint temporal amplitude is obtained by taking the Fourier transform of the joint spectral amplitude,

\[
f_{\text{franson}}(t_s, t_i) = \int d\omega_l d\omega_s F_{\text{franson}}(\omega_i, \omega_s) e^{i\omega_i t_i} e^{i\omega_s t_s} \tag{13}
\]

from which we can obtain the joint temporal intensity,

\[
|f_{\text{franson}}(t_s, t_i)|^2 \propto f_{ss}^2(t_s, t_i) + f_{ds}^2(t_s, t_i) + f_{dl}^2(t_s, t_i) + f_{ll}^2(t_s, t_i) + 2[f_{ss}(t_s, t_i) f_{ls}(t_s, t_i) + f_{sl}(t_s, t_i) f_{ll}(t_s, t_i)] \cos (\omega_0 \tau_s - \phi_s) \\
+ 2[f_{ss}(t_s, t_i) f_{sl}(t_s, t_i) + f_{ls}(t_s, t_i) f_{ll}(t_s, t_i)] \cos (\omega_0 \tau_l - \phi_l) \\
+ 2[f_{sl}(t_s, t_i) f_{ls}(t_s, t_i) \cos (\omega_0 \tau_s - \phi_s) - (\omega_0 \tau_l - \phi_l)] \\
+ 2f_{ss}(t_s, t_i) f_{ll}(t_s, t_i) \cos [(\omega_0 \tau_s - \phi_s) + (\omega_0 \tau_l - \phi_l)], \tag{14}
\]

where

\[
f_{ss}(t_s, t_i) = \exp \left[ - (\sigma_{\omega_s} t_s - \sigma_{\omega_i} t_i)^2 - 2(1 + \rho) \sigma_{\omega_s} \sigma_{\omega_i} t_s t_i \right] \tag{15}
\]

\[
f_{ls}(t_s, t_i) = \exp \left[ - (\sigma_{\omega_s} (t_s + \tau_s) - \sigma_{\omega_i} t_i)^2 - 2(1 + \rho) \sigma_{\omega_s} \sigma_{\omega_i} (t_s + \tau_s) t_i \right] \tag{16}
\]

\[
f_{sl}(t_s, t_i) = \exp \left[ - (\sigma_{\omega_s} t_s - \sigma_{\omega_i} (t_i + \tau_i))^2 - 2(1 + \rho) \sigma_{\omega_s} \sigma_{\omega_i} t_s (t_i + \tau_i) \right] \tag{17}
\]

\[
f_{ll}(t_s, t_i) = \exp \left[ - (\sigma_{\omega_s} (t_s + \tau_s) - \sigma_{\omega_i} (t_i + \tau_i))^2 - 2(1 + \rho) \sigma_{\omega_s} \sigma_{\omega_i} (t_s + \tau_s)(t_i + \tau_i) \right] \tag{18}
\]

are the four terms that represent the different combinations of paths the photons can take in the Franson interferometer, either short-short \( (f_{ss}) \), long-short \( (f_{ls}) \), short-long \( (f_{sl}) \), or long-long \( (f_{ll}) \). These are two-dimensional correlated Gaussian functions that are shifted with respect to the origin by the applied delays \( \tau_s \) and \( \tau_l \). Different types of interference can occur between these paths. The first line in Eq.\( \text{[14]} \) contains the non-interference terms, the second and third lines accounts for single-photon interference, while the fourth and fifth lines account for nonlocal two-photon interference, which depends on the overlap between \( f_{ls} \) and \( f_{sl} \) and between \( f_{ss} \) and \( f_{ll} \), respectively. For anti-correlated photons \( (\rho \rightarrow 1) \), the short-long \( f_{sl} \) and long-short \( f_{ls} \) terms do not overlap and the fourth line goes to zero since \( f_{sl} f_{ls} \rightarrow 0 \). The single-photon temporal marginal, on the other hand, is given by,

\[
|f_{\text{marginal}}(t)|^2 \propto \exp (-2t^2 (1 - \rho^2) \sigma_{\omega}^2) + \exp \left( -2(t + \tau)^2 (1 - \rho^2) \sigma_{\omega}^2 \right) \\
+ 2 \exp \left( -2t^2 (1 - \rho^2) \sigma_{\omega}^2 (t + \frac{\tau}{2})^2 - \frac{2}{2} \sigma_{\omega}^2 \tau^2 \right) \cos (\tau \omega_0 - \phi). \tag{19}
\]
Comparing Eq. 14 and Eq. 19 we find, as before, two different timescale for two-photon and single-photon interference. The interference term which varies as \( \phi_s + \phi_i \) depends on the overlap between \( f_{ss} \) and \( f_{ll} \), whereas the single-photon interference has a coherence time that depends on the inverse bandwidth \( (1/\sigma_\omega = 1/\Delta \omega) \) of the downconverted light.

In order to calculate the expected coincidence and single-photon rates with temporal selection, we consider the limiting case where we temporally select only the photon arrival times halfway between the short and long paths. This is equivalent to setting \( t_s = -\tau_s/2 \) and \( t_i = -\tau_i/2 \) in Eq. 14 and Eq. 19, which simplify to,

\[
\left| f_{\text{franson}} \left( -\frac{\tau_s}{2}, -\frac{\tau_i}{2} \right) \right|^2 \propto \frac{2}{\Delta \omega} \left[ \cos \left( \omega_0 \tau_s - \phi_s \right) + \cos \left( \omega_0 \tau_i - \phi_i \right) \right]
\]

\[ + \exp \left[ -\frac{1}{2} \left( \sigma_\omega \tau_s - \sigma_\omega \tau_i \right)^2 \right] \left[ 1 + \cos \left( \omega_0 \tau_s + \omega_0 \tau_i - \phi_s - \phi_i \right) \right] \]

\[ \left| f_{\text{marginal}} \left( -\frac{\tau_s}{2} \right) \right|^2 \propto \exp \left[ -\frac{1}{2} \left( 1 - \rho \omega \right)^2 \sigma_\omega^2 \tau^2 \right] \left[ 1 + \cos \left( \omega_0 \tau - \phi_j \right) \right] \]

The visibility of the two-photon interference term in Eq 20 is then maximized under two conditions: the ratio of the delays is proportional to the ratio of the marginal bandwidths, \( \sigma_\omega \tau_s = \sigma_\omega \tau_i \), and the delays are less than the two-photon coherence time, \( \tau_s \tau_i \ll 1/[2(1+\rho)\sigma_\omega \sigma_\omega] \). Under these conditions, assuming the photon bandwidths are equal, and substituting the expressions for the single- and two-photon spectral bandwidths, the coincidence rate and single-photon rates of photon detections with temporal selection at \( t_i = -\tau_i/2 \) and \( t_s = -\tau_s/2 \) become,

\[
C(\phi_s, \phi_i) = \left| f_{\text{franson}} \left( -\frac{\tau_s}{2}, -\frac{\tau_i}{2} \right) \right|^2 \propto \exp \left( -\frac{1}{2} \Delta \omega \left( \omega_s + \omega_i \right)^2 \tau^2 \right) \left[ 1 + \cos \left( \omega_0 \tau_s + \omega_0 \tau_i - \phi_s - \phi_i \right) \right]
\]

\[
S(\phi_j) = \left| f_{\text{marginal}} \left( -\frac{\tau}{2} \right) \right|^2 \propto \exp \left( -\frac{1}{2} \left( 1 - \rho \omega \right)^2 \Delta \omega^2 \tau^2 \right) \left[ 1 + \cos \left( \omega_0 \tau - \phi_j \right) \right]
\]

As before, single-photon interference is removed by making the delays larger than the single-photon coherence time, \( \tau \gg 1/\Delta \omega \). However now, with temporal selection, the non-interfering terms have been filtered and 100% interference visibility can be achieved in the two-photon coincidence rate.