Reconstructing ultrafast energy-time entangled two-photon pulses

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The generation of ultrafast laser pulses and the reconstruction of their electric fields is essential for many applications in modern optics. Quantum optical fields can also be generated on ultrafast time scales, however, the tools and methods available for strong laser pulses are not appropriate for measuring the properties of weak, possibly entangled pulses. Here, we demonstrate a method to reconstruct the joint-spectral amplitude of a two-photon energy-time entangled state from joint measurements of the frequencies and arrival times of the photons, and the correlations between them. Our reconstruction method is based on a modified Gerchberg-Saxton algorithm. Such techniques are essential to measure and control the shape of ultrafast entangled photon pulses.

Introduction—The generation, control, and measurement of high-dimensional entangled quantum states of light are important for optical computing and communication [1–4]. One form of this entanglement, in the energy-time degree of freedom, can exhibit strong correlations in frequency and time [5–7], nonlocal interference phenomena [7,8], and time intensity correlations of energy-time entangled [34–36]. To measure both the frequency has also led to nonlinear ultrafast approaches for characterizations [34–36]. The increased interest in time-frequency modes [25–30], or two-photon interference effects [31–33], the only thing fast enough to measure an ultrafast laser pulse. Techniques such as FROG [23] and SPIDER [24] make use of nonlinear optical processes to measure and reconstruct ultrafast pulses. However, adapting them to quantum states of light is challenging due to the low power levels of single photons. In addition, the algorithms developed for laser pulses do not account for the possibility that photons can be entangled. New innovations are therefore needed to reconstruct the joint state of entangled ultrafast photon pulses.

In ultrafast optics and laser physics, the ability to measure the amplitude and phase of laser pulses on ultrafast timescales is essential for nonlinear optics and spectroscopy. In this context, the problem of electric field reconstruction has been extensively studied [21]. Optical pulses can be produced on time scales much shorter than any photodetector response time [22], and consequently, the only thing fast enough to measure an ultrafast laser pulse is another ultrafast pulse. Techniques such as FROG [23] and SPIDER [24] make use of nonlinear optical processes to measure and reconstruct ultrafast pulses. However, adapting them to quantum states of light is challenging due to the low power levels of single photons. In addition, the algorithms developed for laser pulses do not account for the possibility that photons can be entangled. New innovations are therefore needed to reconstruct the joint state of entangled ultrafast photon pulses.

Approaches for characterizing the optical modes of photons have been explored using homodyne measurements [25–30], or two-photon interference effects [31–33]. The increased interest in time-frequency modes has also led to nonlinear ultrafast approaches for characterization [34–36]. To measure both the frequency and time intensity correlations of energy-time entangled states, optical methods based on optical gating and frequency resolved measurements have recently been developed. These have been used to observe nonlocal dispersion cancellation [37] and two-photon quantum interference [38] on time scales inaccessible to standard photodetectors. For complete characterization, however, the joint spectral phase is also required.

Recovering the phase of a field from intensity measurements in Fourier-related domains is known as a phase-retrieval problem. In 1972, Gerchberg and Saxton provided a practical solution to this problem. They introduced an iterative algorithm, referred to as the Gerchberg-Saxton algorithm (GS), to extract the complete wavefunction of an electron beam, including its phase, from intensity recordings in the image and diffraction planes [39]. Their algorithm can be applied to problems involving electromagnetic waves [40] including optical wavelengths [12].

In this Letter, we implement a technique to recover the phase of ultrafast energy-time entangled two-photon pulses based on intensity measurements of the frequency and the arrival time. Inspired by the conventional phase retrieval problem, we develop an algorithm based on a method of alternate projections [39] [43,44] that iterates between the frequency and time domains imposing the measured intensity constraints at each iteration. Measurements in frequency are performed with single-photon spectrometers and measurements in time are implemented via optical gating with an ultrafast optical laser pulse.

Theory—A pure energy-time two photon state can be modelled as [6,17].

$$|\psi\rangle = \int d\omega_s d\omega_i F_{\omega\omega}(\omega_s, \omega_i) a_s^\dagger(\omega_s) a_i^\dagger(\omega_i)|0\rangle,$$ (1)

corresponding to a superposition of frequency modes for the signal $a_s^\dagger(\omega_s)$ and the idler $a_i^\dagger(\omega_i)$ weighted by the joint spectral amplitude (JSA) function $F_{\omega\omega}(\omega_s, \omega_i)$. The joint spectral amplitude, $F_{\omega\omega}(\omega_s, \omega_i) = |F_{\omega\omega}(\omega_s, \omega_i)| \exp[i\phi(\omega_s, \omega_i)]$, describes the amplitude, $|F_{\omega\omega}(\omega_s, \omega_i)|$, and phase, $\phi(\omega_s, \omega_i)$, of the state. For downconversion, it is related to the pump properties
FIG. 1. Block diagram of the algorithm for phase retrieval of an energy-time entangled two-photon state. The algorithm is seeded with an initial guess of the state. At every iteration, the Fast Fourier Transform is applied to one axis of the state after which the magnitude of the state is replaced with the measured data while the phase of the state is preserved. At each iteration the error between the measured and recovered intensities either remains the same or is reduced.

and the phase matching conditions of the nonlinear material [35]. In this form, the joint-spectral intensity $I(\omega_s, \omega_i) = |F_{\omega\omega}(\omega_s, \omega_i)|^2$ characterizes the frequency correlations and the joint temporal intensity (JTI), $I(t_s, t_i) = |F_{tt}(t_s, t_i)|^2$, obtained from the modulus of the Fourier transform, characterizes the temporal correlations. The intensity time-frequency correlations, $I(t_s, \omega_i) = |F_{it}(t_s, \omega_i)|^2$ and $I(t_s, \omega_i) = |F_{it}(t_s, \omega_i)|^2$, can provide additional information on the spectral phase for entangled states [37].

The phase retrieval algorithm is shown in Fig. 1. Four time-frequency intensity correlation measurements are performed, $I(\omega_s, \omega_i), I(t_s, \omega_i), I(\omega_s, t_i), I(t_s, t_i)$ [37]. The effect of the limited instrument resolution for each measured intensity is deconvolved using a Wiener Filter [46]. The algorithm is seeded with an initial guess accompanied with a random phase. In the first iteration, we project the state onto the constraint set that satisfies the measured intensities in frequency. This is achieved by replacing the spectral amplitudes $|F_{\omega\omega}(\omega_s, \omega_i)|$ with the measured spectral amplitudes $\sqrt{I(\omega_s, \omega_i)}$ but keeping the phase,

$$F_{\omega\omega}(\omega_s, \omega_i) \rightarrow \frac{F_{\omega\omega}(\omega_s, \omega_i)}{|F_{\omega\omega}(\omega_s, \omega_i)|} \sqrt{I(\omega_s, \omega_i)}.$$  \hspace{1cm} (2)

We then apply the Fast Fourier Transform algorithm (FFT) to obtain an estimate of $F_{\omega\omega}(t_s, \omega_i)$ and again replace the amplitudes $|F_{\omega\omega}(t_s, \omega_i)|$ with the measured amplitudes $\sqrt{I(t_s, \omega_i)}$. This is repeated two more times, as in Fig. 1, completing one iteration of the algorithm. At each iteration, we evaluate the FROG-trace error [37] between the measured and the reconstructed joint spectral intensities, which corresponds to the average percentage error in each point $(\omega_s, \omega_i)$. An important feature of these types of algorithms is that the measured error will always decrease or remain constant at each iteration, and will not diverge [39, 48].

Phase retrieval algorithms have a well known ambiguity. If the intensity distribution in the Fourier plane is centro-symmetric, then the complex conjugate of any given solution in the object plane is also a solution [39]. For the energy-time degree of freedom, this implies a time-reversal ambiguity, i.e., it is not possible to distinguish between positive and negative dispersion from the intensity measurements in frequency and time alone. Measurements of the time-frequency correlations can distinguish between these two cases and break the time-reversal ambiguity. We find a significant improvement of the algorithm’s performance when these are included in the constraint set.

**Experiment**—The setup is schematically depicted in Fig. 2 and described in detail in Refs. [37, 38]. We produce pairs of energy-time entangled photons at 823 nm and 732 nm using spontaneous parametric downconversion. These are coupled in single-mode fibres allowing for direct, spectrally resolved, or temporally resolved measurements. Spectral measurement are performed via monochrometers with a resolution of 0.1 nm. Temporal
measurements are implemented via optical gating, i.e., via noncollinear sum-frequency generation (SFG) with femtosecond laser pulses in 1 mm of bismuth borate (BiBO) crystal. The electric field of the gate pulse is characterized using an SHG-FROG measurement, and we find an intensity pulse width of 130 fs (s.d.). The instrument resolutions set the filter functions used in the numerical deconvolution of the measured intensities. The quadratic spectral phase on the photons, \( \phi(\omega_s, \omega_i) \approx A_s (\omega_s - \omega_{s0})^2 + A_i (\omega_i - \omega_{i0})^2 \), is controlled with a combination of normally dispersive single-mode fibre and adjustable grating compressor for anomalous dispersion \([37]\), where \( A_s \) and \( A_i \) are the chirp parameters for the signal and idler, respectively. The relative position of the gratings inside the compressor sets the magnitude and sign of the overall dispersion. We calibrate both grating compressors using XFROG (Cross-correlation FROG) spectrogram measurements between the strong gate pulse and a weak laser pulse. The weak laser pulse has the same centre wavelength and path through the fibre-compressor system as the photons on each side. The phase at each relative grating separation is reconstructed using using the Principal Component Generalized Projection (PCGP) FROG algorithm \([47, 50]\). We find a quadratic phase that depends linearly on the grating separation with slopes of \((-1360 \pm 60) \text{ fs}^2/\text{mm} \) and \((-2190 \pm 70) \text{ fs}^2/\text{mm} \) for the signal and idler respectively. The difference between the two is attributed to the cubic dependence on wavelength of dispersion in a grating compressor \([51]\).

Phase reconstructions—We compare the phase retrieval algorithm on measured data for two-photon states with different amounts of dispersion. We set the grating compressors on the signal and idler side to study four cases: no additional dispersion, with extra positive dispersion applied to the idler, with extra negative dispersion applied to the signal, and with extra negative dispersion applied on both sides. For the case of a two-photon energy-time entangled state with negative dispersion applied to both photons, an example of the four combinations of time and frequency measurements is shown in Fig. 3. Background subtraction, a Wiener Filter, and low-pass filters are applied in Fig. 3 and prior to the
reconstruction \cite{52}. We observe strong anti-correlations in the joint spectral intensity [Fig. 3(a)], however, the joint temporal intensity [Fig. 3(d)] is uncorrelated due to the presence of dispersion on both photons. The observed shears in both the time-frequency intensity plots [Fig. 3(b–c)] also illustrate the presence of negative dispersion. We input these intensity constraints into the phase retrieval algorithm and run the algorithm for 1000 iterations, a number found heuristically after which no reduction in the FROG-trace error is observed. The intensity of the reconstructed wavefunction in frequency and time are shown in Fig. 4. The reconstructed intensities are compared to the measured data from Fig. 3(a) and Fig. 3(d). We find a FROG-trace error between the post-processed and reconstructed spectral intensities after 1000 iterations to be (3.64 ± 0.07)% for the joint spectral intensity and (7.01 ± 0.35)% for the joint temporal intensity.

Note that the marginal bandwidths of the joint spectral intensity in the reconstruction [Fig. 3(a)] are shorter than in the original data [Fig. 3(a)]. Numerical simulations suggest this arises as a result of the phase-matching bandwidth in the optical gating. The effect of the phase mismatch on the reconstruction of two-photon states with optical gating is modeled in the supplementary information.

Figure 5 shows the reconstructed joint spectral phase for the four different cases. Starting with the case where we attempted to minimize the unbalanced dispersion [Fig. 5(a)], we observe a relatively flat spectral phase. We then apply \( A_1 = (0.026 \pm 0.002) \text{ ps}^2 \) of dispersion on the signal photon [Fig. 5(b)], and we observe a positive quadratic variation in the phase along the signal (y) axis, modulo 2\( \pi \), with little variations along the idler (x) axis. When we apply \( A_2 = (-0.025 \pm 0.002) \text{ ps}^2 \) of dispersion to the idler photon [Fig. 5(c)], we observe a negative quadratic variation in the spectral phase along the idler (x) axis, with again little variations along the signal (y) axis. When we apply \( A_3 = (-0.036 \pm 0.003) \text{ ps}^2 \) and \( A_4 = (-0.043 \pm 0.002) \text{ ps}^2 \) of dispersion to the signal and idler [Fig. 5(d)], we observe a negative quadratic variation along the diagonal x-y axis.

For the three cases where dispersion is applied, we fit the reconstructed quadratic spectral phase in Fig. 5(b–d). For each, we unwrap the 2D phase and perform a polynomial fit to the phase distribution. For the reconstruction in Fig. 5(b) we obtain a quadratic spectral phase on the signal of \( A_s = (0.024 \pm 0.003) \text{ ps}^2 \) and for the one in Fig. 5(c), we obtain a quadratic phase on the idler of \( A_i = (-0.026 \pm 0.003) \text{ ps}^2 \). For the reconstruction in Fig. 5(d), we obtain a quadratic phase on the signal and idler of \( A_s = (-0.036 \pm 0.004) \text{ ps}^2 \) and \( A_i = (-0.028 \pm 0.003) \text{ ps}^2 \), respectively. The corresponding uncertainties are obtained from the variance in the fitted spectral phase after performing Monte Carlo simulations assuming Poissonian noise. When dispersion is applied to only one photon, Fig. 5(b) and Fig. 5(c), the phase obtained using the phase-retrieval algorithm corresponds to the reconstructed phases measured using the XFROG algorithm. In the last case, Fig. 5(d), we find a discrepancy between the two. This, again, is likely due to the effect of the phase mismatch on the temporal measurements and on the subsequent reconstruction of two-photon states, which will be more pronounced for the photons which have much larger bandwidth than for the weak pulse used for the XFROG reconstructions (see Supplementary information).

**Conclusion**— We have demonstrated a method to recover ultrafast two-photon energy-time entangled pulses. Our technique is based on a method of alternate pro-
jects that iterates between the frequency and time domains imposing the measured intensity constraints at each iteration. The use of nonlinear phenomena, i.e., optical gating, to measure the timing correlations is an artifact of the time scales at play and is not a fundamental requirement. For sufficiently long pulses, there may exist photodetectors that can measure the temporal intensity directly. For subpicosecond resolution involving optical gating, the effect of phase-matching in the upconversion could be reduced using shorter crystals or angle-dithering. Moreover, extensions of this algorithm to characterize two-photon mixed states may be possible based on techniques used to reconstruct partially coherent light, removing assumptions about the purity of the quantum states. Measurement and reconstruction capabilities similar to those available in ultrafast optics will be essential for developing new applications in quantum state engineering and ultrafast shaping of entangled photons, paving the way to characterizing and manipulating high-dimensional quantum states of light.

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SUPPLEMENTARY INFORMATION

Phase retrieval algorithm

The steps involved in the iterative phase retrieval algorithm for energy-time entangled states. Measured intensities in frequency and time are first deconvolved using a Wiener Filter. The algorithm is seeded with an initial guess of the field, which can consist of the measured amplitudes with a random phase. Steps (1-8) are used when all four intensity constraints are applied.

1. Replace the magnitude of \(F_{\omega s}(\omega_s, \omega_i)\) with the measured values \(F'_{\omega s}(\omega_s, \omega_i) = \frac{F_{\omega s}(\omega_s, \omega_i)}{|F_{\omega s}(\omega_s, \omega_i)|} \sqrt{I(\omega_s, \omega_i)}\)

2. Inverse Fourier transform \(F_{\omega s}(\omega_s, \omega_i)\) to obtain an estimate of \(F_{ts}(t_s, \omega_i)\)

3. Replace the magnitude of \(F_{ts}(t_s, \omega_i)\) with the measured values \(F'_{ts}(t_s, \omega_i) = \frac{F_{ts}(t_s, \omega_i)}{|F_{ts}(t_s, \omega_i)|} \sqrt{I(t_s, \omega_i)}\)

4. Inverse Fourier transform \(F'_{ts}(t_s, \omega_i)\) to obtain an estimate of \(F_{tt}(t_s, t_i)\)

5. Replace the magnitude of \(F_{tt}(t_s, t_i)\) with the measured values \(F'_{tt}(t_s, t_i) = \frac{F_{tt}(t_s, t_i)}{|F_{tt}(t_s, t_i)|} \sqrt{I(t_s, t_i)}\)

6. Fourier transform \(F'_{tt}(t_s, t_i)\) to obtain an estimate of \(F_{wt}(\omega_s, t_i)\)

7. Replace the magnitude of \(F_{wt}(\omega_s, t_i)\) with the measured values \(F'_{wt}(\omega_s, t_i) = \frac{F_{wt}(\omega_s, t_i)}{|F_{wt}(\omega_s, t_i)|} \sqrt{I(\omega_s, t_i)}\)

8. Fourier transform \(F'_{wt}(\omega_s, t_i)\) to obtain an estimate of \(F_{\omega s}(\omega_s, \omega_i)\)

Reconstructing two-photon states with optical gating

We construct a numerical model of optical gating and consider its effect on the measurement of two-photon states and the wavefunction reconstruction. The initial state is a two-photon energy-time entangled state as in Eq. 1 with a joint-spectral amplitude for the signal \(\omega_s\) and idler \(\omega_i\) frequencies modeled as a two-dimensional correlated Gaussian function, \(F_{\omega s}(\omega_s, \omega_i)\).

\[
F_{\omega s}(\omega_s, \omega_i) = \frac{1}{\sqrt{2\pi \sigma_{\omega_i} \sigma_{\omega_s} (1 - \rho_{\omega}^2)^{1/4}}} \exp \left(-\frac{1}{2 (1 - \rho_{\omega}^2)} \left[ \frac{(\omega_s - \omega_{\omega s})^2}{2\sigma_{\omega_s}^2} + \frac{(\omega_i - \omega_{\omega i})^2}{2\sigma_{\omega_i}^2} - \frac{\rho_{\omega} (\omega_s - \omega_{\omega s})(\omega_i - \omega_{\omega i})}{\sigma_{\omega_s} \sigma_{\omega_i}} \right] \right). \tag{3}
\]

The marginal frequency bandwidths, \(\sigma_{\omega_s}\) and \(\sigma_{\omega_i}\), and statistical correlation, \(\rho_{\omega}\), of the state are set to the values measured experimentally. We model the optical gating as sum-frequency generation process in the low-efficiency regime between the photons on each side and a gate pulse with centre frequency \(\omega_g\) and a pulse duration of 0.130 ps,
leading to upconverted frequencies $\omega_{us} = \omega_s + \omega_g$ and $\omega_{ui} = \omega_i + \omega_g$ on the signal and idler side, respectively. The gate pulse is modeled with a Gaussian temporal profile,
\[
G(\omega_g, \tau_g) = \frac{1}{(2\pi \sigma_g^2)^{\frac{1}{4}}} \exp\left( -\frac{(\omega_g - \omega_{g0})^2}{4\sigma_g^2} + i\tau_g (\omega_g - \omega_{g0}) \right),
\]
with marginal bandwidth $\sigma_g$, and delay $\tau_g$. The three intensity measurements involving optical gating are calculated via the following,
\[
I(\tau_s, \omega_i) = \int d\omega_s \int d\omega_i G(\omega_{us} - \omega_s, \tau_s) \Phi_{\text{SFG}}(\omega_s, \omega_{us} - \omega_s, \omega_{us}) F_{\omega\omega}(\omega_s, \omega_i)^2,
\]
\[
I(\omega_s, \tau_i) = \int d\omega_i \int d\omega_i G(\omega_{ui} - \omega_i, \tau_i) \Phi_{\text{SFG}}(\omega_i, \omega_{ui} - \omega_i, \omega_{ui}) F_{\omega\omega}(\omega_s, \omega_i)^2,
\]
\[
I(\tau_s, \tau_i) = \int d\omega_{ui} \int d\omega_{ui} G(\omega_{us} - \omega_s, \tau_s) \Phi_{\text{SFG}}(\omega_s, \omega_{us} - \omega_s, \omega_{us}) \times G(\omega_{ui} - \omega_i, \tau_i) \Phi_{\text{SFG}}(\omega_i, \omega_{ui} - \omega_i, \omega_{ui}) F_{\omega\omega}(\omega_s, \omega_i)^2
\]
where the gate pulse $G$ is the same on both sides but with delays $\tau_i$ and $\tau_s$ introduced. The phase matching function is,
\[
\Phi_{\text{SFG}}(\omega_j, \omega_{uj} - \omega_j, \omega_{uj}) = \exp\left( -i\frac{\Delta k L}{2} \right) \text{sinc}\left( \frac{\Delta k L}{2} \right)
\]
where the phase-mismatch,
\[
\Delta k(\omega_j, \omega_{uj} - \omega_j, \omega_{uj}) = \frac{n_c(\omega_j) \omega_j}{c} + \frac{n_c(\omega_{uj} - \omega_j)(\omega_{uj} - \omega_j)}{c} + \frac{n_o(\omega_{uj}) \omega_{uj}}{c}
\]
is calculated for type-I SFG with different crystal lengths $L$ and the experimentally measured wavelengths. The phase-matching bandwidth can be estimated from the range of frequencies contained in $\Delta k L = \pi$. Upconverted frequencies outside this range are suppressed. All integrals are evaluated numerically.

In the SFG process used for optical gating, a photon and a strong gate pulse in the near-infrared (NIR) are up-converted to produce a higher energy photon in the ultraviolet. If the photons are dispersed before the optical gating, high and low frequencies components will arrive at different times in the nonlinear medium. In the presence of phase mismatch, the upconverted frequencies associated to these high and low frequency components can lie outside the phase-matching bandwidth of the crystal, and consequently will be suppressed. As a result, phase mismatch in optical gating changes the measured intensity correlations and therefore changes the intensity constraints that are applied in the phase retrieval algorithm. The applied constraints will no longer correspond to the modulus of the Fourier transform of a physical state, thereby affecting the reconstruction.

We model all the steps in the phase-retrieval process. We numerically create frequency anti-correlated states, with the same centre wavelength and bandwidth as those measured experimentally, but with different amounts of applied spectral phases, given by the chirp parameters $A_s$ and $A_i$. We calculate the four joint correlations in frequency and time using different lengths of BiBO for optical gating, apply the numerical deconvolution to each intensity measurement, and insert these as constraints for the phase retrieval algorithm. After reconstruction, we unwrap the spectral phase of the reconstructed joint spectral amplitude function and fit it to a third-order two-dimensional polynomial.

The reconstructed spectral phases are compared to the applied spectral phases in Fig. 6 for different lengths of BiBO used in optical gating and for different applied spectral phases. In Fig. 6(a) and Fig. 6(b), the signal chirp parameter $A_s$ is kept fixed while the idler chirp parameter is varied, whereas in Fig. 6(c) and Fig. 6(d), the idler chirp parameter $A_i$ is kept fixed while the signal chirp parameter $A_s$ is varied. When the length of the crystal is set to zero ($L = 0 \mu m$), the reconstructed phase corresponds exactly to the applied phase, and the line at $L = 0 \mu m$ appears at 45 degrees with a slope of one. As the length of the crystal increases, we find that the slope remains fairly constant at 45 degrees, but the offset depends on the configuration. For example, comparing Fig. 6(a) and Fig. 6(b), we find the values of the reconstructed idler chirp parameter $A_i$ depend on whether the signal chirp parameter has a value of $A_s = 5,000 \text{ fs}^2$ [Fig. 6(a)] or $A_s = 40,000 \text{ fs}^2$ [Fig. 6]. The difference between the reconstructed and applied phase in Fig. 6 also becomes larger for longer crystals where the phase matching function is more restrictive.
FIG. 6. Effect of phase mismatch on the reconstructed spectral phase. We model the effect of optical gating with different lengths $L$ of BiBO on the reconstructed phase. The reconstructed phase is compared to the applied phase for four different cases. The signal chirp parameter is fixed to the values of (a) $A_s = 5,000 \text{ fs}^2$ and (b) $A_s = 40,000 \text{ fs}^2$ while the idler chirp parameter $A_i$ is varied. The idler chirp parameter is fixed to the values of (c) $A_i = 5,000 \text{ fs}^2$ and (d) $A_i = 40,000 \text{ fs}^2$ while the signal chirp parameter $A_s$ is varied. At $L = 0 \mu m$, the reconstructed phase is the same as the applied phase. As $L$ is increased, phase mismatch becomes more important and this changes the value of the reconstructed phase.