Octave-spanning spectral phase control for single-cycle bi-photons

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Abstract
The quantum correlation of octave-spanning time–energy entangled bi-photons can be as short as a single optical cycle. Many experiments designed to explore and exploit this correlation require a uniform spectral phase (transform-limited) with very low loss. So far, transform-limited single-cycle bi-photons have not been demonstrated, primarily due to the lack of precise, broadband control of their spectral phase. Here, we demonstrate the correction of the spectral-phase of near-octave spanning bi-photons to $20\phi_\pi$ over an octave in frequency ($1330\approx2600$ nm). Using a prism-pair with an effectively negative separation for shaping the bi-photons’ spectral phase, we obtain a tuned, very low-loss compensation of both the second and fourth dispersion orders. An essential requisite for precise tuning over such a broad bandwidth is a measure of the spectral phase that provides feedback for the tuning even when the overall dispersion is far from compensated. This is achieved by a non-classical bi-photon interference, which enables direct verification of the corrected bi-photon spectral phase.

1. Introduction
The ultra-broad bandwidth of time–energy entangled photons that are generated from a narrowband pump (bi-photons) enfold a unique resource for quantum information. Due to the ultra-short temporal correlation, yet continuous generation, an ultra-high flux of single bi-photons can be generated, which is specifically attractive for high-speed quantum communication and quantum cryptography [1–3]. Single-cycle bi-photons, whose bandwidth covers an octave in frequency and temporal correlation is comparable to the optical cycle, are the extreme realization of time–energy entanglement, and a long-sought goal in experimental quantum optics [4–6]. In the pursuit of single-cycle bi-photons, two main limitations need to be overcome. First, the phase matching condition should be broad enough to allow bi-photon generation with sufficient bandwidth; and second, precise spectral-phase control is necessary to overcome the dispersive temporal broadening of the correlation and to compress it to a single-cycle duration. In addition, low loss is important, as the non-classical correlation between the photons is quadratically sensitive to photon loss.

The main difficulty in generating broadband bi-photons collinearly is how to achieve ultra-broad phase matching of the down-conversion process in the nonlinear medium. Two approaches exist in this respect. One approach is the spatial variation of the phase matching conditions within the crystal (chirping the polling period) to accommodate broadband quasi-phase matching. Another approach is to choose a generation wavelength that allows compensation of the entire bandwidth by a single phase-matching condition, which is possible when the central bi-photon frequency is matched to the zero dispersion of the nonlinear medium. Ultra-broad phase matching of the nonlinear down-conversion interaction, has been widely discussed [4–10]. In [6, 9, 10] for example, the broad bandwidth is achieved by chirping the quasi-phase matching period along the crystal. We have recently demonstrated generation of near-octave, ultra-broad bi-photons using type-I down-conversion near the zero-dispersion frequency of the nonlinear crystal [7, 8].

Precise control of the spectral phase has been far less addressed. When the spectral phase of the bi-photons is modulated by group velocity dispersion (GVD) in matter, or by any other means, the bi-photons are no longer transform-limited and the major bi-photon measurement methods, Hong–Ou–Mandel (HOM) interference...
and frequency generation (SFG) [2, 4, 14–16] are no longer applicable. The reason for this is that both SFG and HOM are broadband interference effects that are sensitive to phase modulation (in somewhat different ways). SFG is affected by the bi-photons’ phase-sum, implying sensitivity to even orders of the dispersion experienced by the bi-photon beam [17]. The common HOM is affected by the phase-difference between two indistinguishable photons, indicating that HOM is sensitive to odd orders of the dispersion-difference between the two interfering beams [13] (interestingly, if the HOM effect is time-reversed [18] it too becomes sensitive to even orders). Compensation of the spectral phase over such an ultra-broad bandwidth is not trivial, as it requires tuned compensation of several orders of dispersion. Besides the need for compensation, a measure of the spectral phase is also necessary as feedback to guide the tuning while the overall dispersion is still far from compensated. Thus, even using the SFG or HOM signals as feedback for compensation tuning is insufficient, and a spectrally-resolved interference effect is required.

To fully exploit the bi-photon bandwidth the spectral phase-shaping mechanism needs to provide independent compensation of at least two orders of dispersion (second and fourth) and to be very low-loss in order to maintain the quantum bi-photon state. In addition, since the bi-photons are generated in many experiments in the IR range, where the dispersion of most optical materials becomes negative, standard techniques for dispersion control from the visible and near-IR range, such as a prism-pair [19] or grating-pair [20] are no longer applicable, since they inherently rely on the negative dispersion of free space between dispersive elements in the bi-photon beam. For comparison we point out a few other common methods for dispersion compensation and examine their suitability for our case. The most basic mechanism is material GVD [21], which offers one non-tunable degree of freedom (length) to compensate one dispersion order. This also depends on the existence of a convenient optical material with GVD of the required sign. Group delay dispersion (GDD) designed dielectric coatings [22], can offer full compensation with proper design and realization, but are costly, not tunable and have a limited dynamic range for phase correction. A spatial light modulators (SLM) [17, 18] is a most general tunable compensation mechanism, although it is lossy and costly with limited spatial resolution and dynamic range. Last, grating-pairs [13] can compensate very large dispersion values yet they tend to be lossy and limited in their ability to compensate two dispersion orders simultaneously.

Here, we describe and demonstrate compensation of the bi-photon spectral phase to <π/20 across a bandwidth of >10 THz (nearly an octave), using a Brewster prism-pair [19] with an effectively negative separation that allows simultaneous compensation of two dispersion orders: second (GDD) and fourth (fourth-order dispersion, FOD). As feedback for tuning the compensation we exploit a pairwise interference effect [8] that can measure the bi-photon spectral phase even when the dispersion is totally uncompensated. This interference measure provides clear verification of the compensation accuracy. Note that only the phase-sum of both photons of the bi-photon pair is defined, which is affected by even orders of dispersion. The phase of the single photons (or phase difference between them), which is affected by odd dispersion orders, remains undefined. In the case of SFG, for example, since both signal and idler photons experience the same dispersion, compensation is needed only for even (symmetric) orders [16, 23–25], while odd (anti-symmetric) orders have no influence, as they keep the phase-sum unaffected. Nonetheless, the compensation method used here can be relevant for odd orders of dispersion also, where necessary.

2. Dispersion control concept

Our broadband bi-photon source relies on spontaneous parametric down-conversion (SPDC), where the bandwidth of the bi-photons is limited by phase matching. Ultra-broadband SPDC is obtained by matching the center frequency of the bi-photon spectrum (half the pump frequency) with the zero-dispersion of the nonlinear crystal. We use a periodically-poled KTP (PPKTP) crystal, pumped by a single-frequency laser at 880 nm. The generated SPDC is symmetrical in frequency around the degenerate wavelength, λ₀ = 1760 nm, which is nearly the zero dispersion wavelength λ = 1790 nm. With this method, we generate bi-photons spanning from ≈115 THz to ≈225 THz around the center (degenerate) frequency of λ₀ = 170 THz. The dispersion that needs compensation is initially the residual phase mismatch in the crystal, in addition to dispersion from optical elements in the bi-photon beam.

A common simple technique for dispersion compensation uses a pair of Brewster- cut prisms [26] as illustrated in figure 1(a). By varying the separation R between the prisms tips and the insertion H of the prisms into the beam path, the geometrical and material dispersion can be dynamically controlled to tune the overall dispersion. The Brewster prism-pair is preferable to other techniques of dispersion control due to its ultra-low loss and high degree of tunability in real time.
In ultrafast applications in the visible and the NIR range, the prism material GDD is positive, whereas the geometrical dispersion created by the separation between the prisms always introduces negative GDD. This technique is therefore most suitable for dispersion compensation in the visible and NIR spectrum, where most optical materials produce positive GDD. In our case, however, the broad bi-photons spectrum is in the short-wavelength IR (SWIR) range and beyond, where most optical materials produce negative GDD. Thus, the separation of the prisms cannot compensate for material dispersion, considerably limiting the choice of materials that can match the experimental needs, and posing a major hurdle for the prism-pair to produce an overall compensation.

In some cases, a delicate balancing of the two dispersion knobs, separation \( R \) and insertion \( H \) of the prisms, enables compensation of two orders of dispersion, such as the second and third orders (GDD and TOD). This is a unique feature of the double-prism (compared to the grating pair for example), that allows ultra-broad phase compensation without an SLM in a phase shaper [18] which is costly, complicated and lossy. However, such a simultaneous compensation of two orders is not guaranteed, as it may require a 'non-physical' configuration with either negative separation between the prisms or a negative insertion of the prism into the beam. Indeed, we found that for compensating both GDD and FOD of our bi-photons, the solution requires a negative separation between the prisms. Note that negative separation can be physical if we introduce an imaging system between the two prisms [27, 28], which images the first prism tip beyond the location of the second prism, as illustrated in figure 1(b).

Obviously, the introduction of a negative separation inverts the sign of the geometrical dispersion and enables the prism-pair to produce a total positive GDD, even for prism material with negative dispersion.

In our experiment, this allows compensation of both GDD and FOD, which is impossible with a standard, positively separated prism-pair or with a grating-pair [20, 29]. Our analysis of the frequency dependent optical path in the prism-pair relies on a generalization of the original method presented by Fork [19] and is detailed in [30].

3. Experiment

We measured experimentally the bi-photon spectral phase with the non-classical interference effect presented in [8], which is capable of measuring the bi-photon spectral phase and amplitude even in the presence of spectral modulation. The SPDC that was generated in one nonlinear crystal propagated together with the pump laser into a second identical crystal, where SPDC can be either enhanced or diminished. Quantum mechanically, the two possibilities to generate bi-photons (either in the first crystal or the second) interfere according to the relative phase between the pump and the SPDC, which was acquired between the crystals. Two types of relative phase are possible: (1) phase of the pump itself acquires a phase relative to the entire bi-photon spectrum, and (2) a spectrally varying phase over the SPDC spectrum. The first leads to a intensity variation of the entire spectrum together, whereas the second leads to the appearance of interference fringes across the spectrum. By analyzing the spectral interferogram, we can reconstruct the spectral phase. When the dispersion is fully

![Figure 1. (a) Standard prism pair configuration: an optical axis is defined along the main beam of a wavelength that is exactly at minimum deviation (matched to the Brewster angle \( \theta_0 \)) through the Brewster cut prisms. \( R \) is the distance between the prisms tips (A,B) along this axis, and \( H \) is the insertion of the prisms into the beam relative to this axis (B, C). Other wavelengths are deviated by a dispersion angle \( \delta \theta \). (b) A prism-pair with negative separation: the telescope images the vertex of the first prism forward (distance 4f). Placing the second prism before the image results in an effective negative separation \( R \) between the prisms.](image)
compensated, the spectral phase becomes flat and only uniform variation of the entire spectrum will be observed without any spectral fluctuation, when the pump phase is varied.

The experimental configuration is illustrated in figure 2 and consists of three main parts: first, ultra broadband bi-photons are generated via collinear SPDC in a Brewster cut 12 mm long, PPKTP crystal pumped by a single-frequency diode laser at 880 nm (of power \( \approx 0.5 \) W). The SPDC produces \( \approx 10^{12} \) bi-photons per second with a bandwidth of \( \approx 100 \) THz. Next, the generated bi-photons propagate through a folded prism-pair system (double-pass), where a pair of sapphire prisms are separated by a reflective telescope constructed from two gold spherical mirrors. By double-passing the prism-pair system, we guarantee exact re-packaging of the spectrum. In our experiment, the negative distance between the tips is a few centimeters, while the prism insertion is several millimeters. The shaped spectrum enters a second identical crystal (along with the pump) where bi-photon interference can occur in the form of enhanced/diminished bi-photon generation (constructive/destructive interference). The generated bi-photon spectrum and spectral fringes are measured with a home-built spectrometer composed of a sapphire prism coupled to a cooled CCD camera. A symmetrical spectral interference pattern is observed, which is then used to extract the spectral phase acquired by the SPDC light between the two crystals (see the appendix for details on the phase-extraction method). The observed interference contrast was rather low (15–20%), primarily due to the use of Brewster cut crystals which introduces a small geometric dispersion of the different frequency components and degraded the spatial-mode matching at the second crystal. Yet, the contrast still enabled high-fidelity extraction of the spectral phase.

We now wish to find the optimal dispersion compensation for the bi-photons among the different possible \((R, H)\) configurations of the prisms (separation and penetration). Optimal compensation should be obtained near the calculated vanishing point of both the GDD and FOD (see figure 3). We therefore varied \( R \) and \( H \) according to the following protocol: we scanned the separation \( R \) from a large negative separation towards \( R = 0 \). For each separation \( R \), we tuned the prism insertion \( H \) to achieve the broadest possible compensation at this \( R \) and estimated the compensated bandwidth by measuring the bandwidth over which the spectral interference variation was uniform (see details later on). Figure 4(a) depicts the measured compensated bandwidth as a function of the negative separation \( R \), showing optimal compensation at \( R \approx -40 \) mm with the prism insertion \( H \approx 14 \) mm. Initially, we aimed for a maximal phase fluctuation of \( \Delta \phi < \pi/10 \) across the entire spectrum, but analysis shows that the compensated fluctuation was better (\( \Delta \phi \approx \pi/20 \)) over the entire >110 THz bandwidth. Figure 4(b) shows the measured spectrum at the optimal point, demonstrating the uniform variation of the entire spectrum between destructive and constructive interference.

The measured residual phase, together with the intensity spectrum of our bi-photons, was used to calculate an estimated SFG correlation time of \( \sim 8 \) fs width (<1.4 optical cycles). The residual phase fluctuation (of zero

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**Figure 2.** Experimental setup: collinear SPDC is generated in the first KTP crystal (marked (a)) and double passes through (b) a prism-pair setup (P1 and P2, sapphire) with an intermediate 4f telescope consisting of two spherical mirrors (M1 and M2, \( f = 500 \) mm). The compensated SPDC spectrum and the 880 nm pump exit at a slightly lower height than the input, and are directed by a lower mirror (HR) below the incoming beam into a second identical KTP crystal (c) where the non-classical interference occurs. The focusing spherical mirrors around the KTP crystals have a focal length of \( f = 75 \) mm. All the mirrors in the setup (spherical and plane) are metallic coated, either silver or gold. The spectrometer consists of a third sapphire prism (P3), a focusing lens \( f = 125 \) mm and a cooled CCD camera (SWIR range).
mean) does not affect the main central lobe of the correlation, but only enhances the correlation side-lobes slightly. However, as long as the residual phase is kept under $\pi/5$ the effect on the side-lobes is practically negligible. The correlation performance of our experimentally corrected spectrum in an SFG measurement should therefore show no noticeable difference compared to an ideal transform-limited spectrum.

4. Conclusion

We demonstrated dispersion compensation of ultra-broadband bi-photons, using a prism-pair with an effectively negative separation, over nearly a full octave. The low-loss ultra-broad phase compensation enables utilization of broadband quantum interference methods, such as HOM and SFG, opening an avenue to quantum optics applications with ultra-broadband, high-flux bi-photons. The compensation quality and bandwidth were verified by a nonlinear pairwise interference, acting as a feedback signal for tuning the compensation.

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Appendix. Extraction of the spectral phase

Let us provide further details on the retrieval of the residual phase fluctuation in the experiment. The spectral interferogram intensity can be expressed as

\[ I_\omega = I_0(\omega)[1 + V(\omega)\cos(\varphi_0 + \varphi(\omega))] \]

where \( I_0(\omega) \) is the average spectrum without interference, \( V(\omega) \) is the fringe visibility at frequency \( \omega \), which varies weakly in our experiment between 15–20\%, \( \varphi(\omega) \) is the spectral phase variation of interest.

In order to retrieve the spectral phase variation, we normalize the interferogram according to

\[ I_{\text{norm}} = \frac{I(\omega)}{I_0(\omega)} - 1 = \frac{V(\omega)\cos(\varphi_0 + \varphi(\omega))}{V_0(\omega)}. \]

The phase variation is most pronounced when the phase of the pump is \( \varphi_0 \approx \pm \pi / 2 \), where \( \cos(\varphi_0 + \varphi(\omega)) \approx \varphi(\omega) \) (assuming a small phase variation \( \varphi(\omega) \)), indicating that the spectral phase is \( \varphi(\omega) = \varphi_{\text{norm}} / V_0(\omega) \), where \( V_0(\omega) \) is the overall phase of the pump.

Figure A1 shows representative sets of normalized interferograms for non-optimal compensation (figure A1(a)) and at the optimal point (figure A1(b)), where the entire spectrum apparently varies uniformly. In order to demonstrate full compensation, we have recorded a large set of normalized interferograms at various \( \varphi_0 \) and analyzed specifically those of \( \varphi_0 \approx \pi / 2 \). For figure A1(b), analysis indicates a residual phase variation of \(<\pi/20\%\).

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