Supplementary Information

Designing High-Power, Octave Spanning Entangled Photon Sources for Quantum Spectroscopy - Supplementary Information

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I. THEORY

A. CLT Refractive Indices

The Sellmeier equations [1] describing the frequency and temperature dependent refractive indices of the material are plotted in Fig.1.

![Fig. 1: Congruent lithium tantalate refractive indices for ordinary and extraordinary polarizations at two different temperatures.](image)

B. Quasi-Phase-Matching

Based upon these values, the poling periodicity was calculated for a collinear 3rd-order Type-0 QPM configuration with a degenerate wavelength of 812nm.

![Fig. 2: calculated poling period for 406nm-812nm at 133°C - CLT](image)
C. Phase-matching Amplitude

Once the required poling period for our specific crystal and configuration has been determined, the two-dimensional phase-matching amplitude can be calculated by again evaluating Eq. 5 and Eq. 7 in the main text for a wide range of wavelength combinations. The result is the contour plot in Fig. 3, which shows the downconversion intensity for photon pairs near the 812 nm degeneracy point. The inset of Fig. 3 is a wide-range plot displaying the characteristic shape of the phase-matching curve for a Type-0 process. The contour provides some important information regarding the resultant output of the SPDC process. Mathematically, the output state of the downconversion process can be described as:

$$|\Psi\rangle = \int \int \gamma(\omega_s, \omega_i) d\omega_s d\omega_i a_s^\dagger a_i^\dagger |0\rangle$$ (1)

whereby the joint spectral amplitude (JSA), $\gamma(\omega_s, \omega_i)$ is the product of two terms, the pump-envelope function $\mu(\omega_s, \omega_i)$ and the phase-matching amplitude $\phi(\omega_s, \omega_i)$ [2]. Here the phase-
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matching amplitude function is solely determined by the crystal and the desired downconversion configuration. The pump-envelope function on the other hand is dictated by, as the name implies, the spectral characteristic of the pump beam. Obviously due to conservation of energy, $\mu$ will be diagonal in its orientation if plotted as a function of the signal and idler wavelengths.

The strongly diagonal direction of $\phi$ is advantageous as its overlap with the pump-envelope along the diagonal allows for a wide range of anti-correlated signal and idler photon pairs to be phase-matched. This is of particular importance in experiments where this anti-correlation is necessary for achieving two-photon absorption processes which occur via an intermediate energy level transition [3]. This can be viewed in contrast to applications where highly spectrally pure photon states are desired, such as in single photon sources. Here, obtaining a lack of frequency correlations by designing an anti-diagonal direction of $\phi$ is of primary concern.

D. Entanglement and Purity

Given the output state as defined in Eq.1, the joint spectral amplitude allows for the prediction of the SPDC spectrum obtained for a given configuration of $\mu$ and $\phi$ by carrying out an integral across all signal/idler wavelengths with respect to a corresponding idler/signal wavelength held constant. For our specific 20 mm long ppCLT grating with a 9.5 $\mu$m poling period, the pump bandwidth has a distinct effect on not only the resulting emission spectrum’s bandwidth, but also more profoundly on the entanglement of the photonic state. This is characterized by the purity,

$$P = \frac{1}{K}$$

(2)

with

$$K = \frac{1}{\sum_i \lambda_i^2}$$

(3)

being the Schmidt number equal to the rank of the reduced single particle density matrix. This value can be interpreted as a weighted measure (weights being the eigenvalues $\lambda_i$) of the number of modes required to represent the mixed state, and therefore how entangled it is [4]. The eigenvalues themselves are obtained via a singular value decomposition of the joint spectral amplitude matrix.

As shown in Fig.4 (top), the SPDC spectrum trivially narrows as the pump bandwidth is decreased up to a limiting point. Below this value, the intersection of the pump envelope $\mu$ with the phase matching amplitude $\phi$ no longer causes a decrease in the spectral width which is being carved out by $\mu$. Fig.4 (bottom) shows the calculated FWHM of the SPDC spectrum as a function of the pump
bandwidth, as well as the corresponding purity of the output state. As is evident, the purity of the state decreases with increasing pump bandwidths, and therefore conversely the entanglement of the overall state increases. While this is advantageous from the perspective of producing a state which displays more/stronger anti-correlations between photon pairs, the significant drawback is that the spectral resolution with which optical atomic & molecular transitions are able to be addressed is directly determined by the bandwidth of the pump laser driving the SPDC process. Thus, for highly spectrally resolved measurements, the figure of merit will be dictated by the pump laser.

FIG. 4: SPDC emission spectrum as a function of the pump bandwidth. The corresponding broadening of the spectrum at larger bandwidths carries with it a degradation of the photon state purity which is of advantage where strong (anti-)correlation between photon pairs is desired

E. Broadband HOM Interference

The temporal properties and the degree of entanglement of the SPDC flux can be assessed by measuring the fourth-order interference in a two-photon Michelson (or Mach-Zehnder) interferometer. In such an interferometer, six beamsplitter ports are involved. See diagram below (Fig.5).
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FIG. 5: Schematics of a two-photon Michelson or Mach-Zehnder interferometer.

The interferometer input wavefunction for entangled pairs entering BS1 via port $a$ is

$$|\Psi\rangle \propto \iint d\omega_s d\omega_t \psi(\omega_s, \omega_t) \hat{a}_s^\dagger(\omega_s)\hat{a}_i^\dagger(\omega_t) |0,0\rangle$$  \hspace{1cm} (4)

where $\psi(\omega_s, \omega_t)$ is the joint spectral amplitude of the entangled photons as defined in previous sections.

After the first pass through BS1,

$$\hat{a}_s^\dagger(\omega_s)\hat{a}_i^\dagger(\omega_t) = \frac{1}{2} [\hat{c}_s^\dagger(\omega_s)\hat{c}_i^\dagger(\omega_t) - \hat{d}_s^\dagger(\omega_s)\hat{d}_i^\dagger(\omega_t) + i\hat{c}_s^\dagger(\omega_s)\hat{d}_i^\dagger(\omega_t) + i\hat{d}_s^\dagger(\omega_s)\hat{c}_i^\dagger(\omega_t)].$$  \hspace{1cm} (5)

After the second pass

$$\hat{a}_s^\dagger(\omega_s)\hat{a}_i^\dagger(\omega_t) = \frac{1}{4} [1 + e^{i\omega_p \tau} - e^{i\omega_s \tau} - e^{i\omega_t \tau}] \hat{c}_s^\dagger(\omega_s)\hat{c}_i^\dagger(\omega_t)$$
$$+ \frac{i}{4} [1 - e^{i\omega_p \tau} + e^{i\omega_s \tau} - e^{i\omega_t \tau}] \hat{d}_s^\dagger(\omega_s)\hat{d}_i^\dagger(\omega_t)$$
$$+ \frac{i}{4} [1 - e^{i\omega_p \tau} - e^{i\omega_s \tau} + e^{i\omega_t \tau}] \hat{d}_s^\dagger(\omega_s)\hat{c}_i^\dagger(\omega_t)$$
$$- \frac{1}{4} [1 + e^{i\omega_p \tau} + e^{i\omega_s \tau} + e^{i\omega_t \tau}] \hat{d}_s^\dagger(\omega_s)\hat{d}_i^\dagger(\omega_t).$$  \hspace{1cm} (6)

Since the coincidences are detected after port $f$, the last term of the above expression is relevant. Using the fact that $\omega_r = \omega_p - \omega_s$, the $ff$ component of the entangled two photon wavefunction becomes

$$|\Psi\rangle_{ff} \propto -\frac{1}{4} \iint d\omega_s d\omega_t \psi(\omega_s, \omega_t) [1 + e^{i\omega_p \tau} (1 + e^{-i\omega_s \tau}) + e^{i\omega_t \tau}] \hat{d}_s^\dagger(\omega_s)\hat{d}_i^\dagger(\omega_t) |0,0\rangle$$
$$= -\sqrt{2} \iint d\omega_s \psi(\omega_s, \omega_p - \omega_s) [1 + \cos((\omega_p - \omega_s) \tau)][1 + \cos(\omega_s \tau)] |0,2\rangle$$  \hspace{1cm} (7)

Note that here it was assumed that $\omega_p$ is constant and the pump laser has an infinitely narrow line width. Therefore, the probability of detecting coincidences after port $f$ is

$$P \propto 2 \iint d\omega_s |\psi(\omega_s, \omega_p - \omega_s)|^2 [1 + \cos((\omega_p - \omega_s) \tau)]^2 [1 + \cos(\omega_s \tau)]^2$$  \hspace{1cm} (8)
where $|\psi(\omega_s, \omega_p - \omega_s)|^2$ is the joint spectral density of the entangled photons. Thus the interference pattern at port $f$ can be plotted out (Fig. 6 and 7). The interference pattern is the result of contributions from both classical and quantum (HOM) interferences. The effect of HOM is manifested by the main peak at zero time delay. As the bandwidth of the entangled photons change, either by varying phase matching conditions or simply placing a bandpass filter before the photon counting detectors, the width of the interference peak, proportional to the coherence length of the flux, changes accordingly.

FIG. 6: Simulated normalized narrowband Michelson interference corresponding to the Gaussian SPDC spectrum filtered by 10 nm bandpass filter centered at 812 nm. Sampling step 0.32 fs.
FIG. 7: Simulated normalized broadband Michelson interference corresponding to the Gaussian SPDC spectrum filtered by 100 nm bandpass filter centered at 812 nm. Sampling step 0.32 fs.

II. ADDITIONAL EXPERIMENTAL DATA

Figs.6-9 provide additional characterization on the relationship between interference measurements and the pump.

FIG. 8: Measured collinear broadband two-photon interference when the arms of the Michelson interferometer are perpendicularly polarized. No distinguishable interference pattern is observed, and the coincidences follow pump and noise fluctuations.
FIG. 9: Measured collinear narrowband two-photon interference when the arms of the Michelson interferometer are perpendicularly polarized. No distinguishable interference pattern is observed, and the coincidences follow pump and noise fluctuations.

FIG. 10: Measured SPAD counts and coincidences vs. pump power in the collinear broadband case. Top: single SPAD channel counts in 15s. Bottom: Total and accidental coincidences in 15s.
FIG. 11: Measured SPAD counts and coincidences vs. pump power in the collinear narrowband case. Top: single SPAD channel counts in 15s. Bottom: Total and accidental coincidences in 15s.
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REFERENCES

1. Moutzouris, K., Hloupis, G., Stavrakas, I., Triantis, D. & Chou, M.-H. Temperature-dependent visible to near-infrared optical properties of 8 mol% Mg-doped lithium tantalate. *Opt. Mater. Express* **1**, 458–465. http://www.osapublishing.org/ome/abstract.cfm?URI=ome-1-3-458 (July 2011).

2. Zielnicki, K. et al. Joint spectral characterization of photon-pair sources. *Journal of Modern Optics* **65**, 1141–1160. eprint: https://doi.org/10.1080/09500340.2018.1437228. https://doi.org/10.1080/09500340.2018.1437228 (2018).

3. Saleh, B. E. A., Jost, B. M., Fei, H.-B. & Teich, M. C. Entangled-Photon Virtual-State Spectroscopy. *Phys. Rev. Lett.* **80**, 3483–3486. https://link.aps.org/doi/10.1103/PhysRevLett.80.3483 (16 Apr. 1998).

4. Ekert, A. & Knight, P. L. Entangled quantum systems and the Schmidt decomposition. *American Journal of Physics* **63**, 415–423. eprint: https://doi.org/10.1119/1.17904. https://doi.org/10.1119/1.17904 (1995).