

Supplementary Information

A. Theoretical Model

Taking into account the finite duration of the pump pulse, the finite length of interaction, and the linearity of energy and momentum conservation, the wave function describing the two-photon state produced by pumping at a given angle θ can be written as :

$$|\Psi(\theta)\rangle = \iiint d\omega_s d\omega_i dz \left[\mathcal{A}_{+\theta}^{(1)}(\omega_s, \omega_i) a_H^\dagger(\omega_s) a_V^\dagger(\omega_i) + \mathcal{A}_{+\theta}^{(2)}(\omega_s, \omega_i) a_V^\dagger(\omega_s) a_H^\dagger(\omega_i) \right] S_+(z) |\text{vac}\rangle |z\rangle \quad (1)$$

where $\mathcal{A}_{+\theta}^{(1)}(\omega_s, \omega_i)$ and $\mathcal{A}_{+\theta}^{(2)}(\omega_s, \omega_i)$ are the joint spectral amplitudes corresponding to the two possible phase-matching conditions, z is the propagation direction of the generated photons and $S_+(z)$ the spatial profile of the pumping beam in the z direction. The functions $\mathcal{A}_{+\theta}^{(1)}(\omega_s, \omega_i)$ and $\mathcal{A}_{+\theta}^{(2)}(\omega_s, \omega_i)$ are completely determined by the pump spectrum and the interaction length in the medium. Notice that, in principle, other photonic degrees of freedom of the pumping beam, as for instance its dispersion in momentum, should be taken into account to provide the most complete theoretical model of the experiment. However, we expect that the spatial and frequency degrees of freedom should play the most important role to explain the observed results, and Fig.5 indicates that our model indeed provides a very good description of the experiment. In general, the two interactions have different central frequencies and since the spectral width is small compared to other types of phase matching, photons generated by interaction (1) are distinguishable from photons generated by interaction (2): frequency provides a “which-way” information on each polarization and thus reduces the entanglement. To obtain indistinguishable photons from both sides of the medium (right and left), two pump laser pulses must be used with different angles of incidence $\pm\theta_{\text{deg}}$. In this case, each pump generates a pair of photons, and the total state can be written as:

$$\begin{aligned} |\Psi(+\theta_{\text{deg}}, -\theta_{\text{deg}})\rangle = & \iiint d\omega_s d\omega_i dz \left[\mathcal{A}_{+\theta_{\text{deg}}}^{(1)}(\omega_s, \omega_i) S_+(z) |\omega_s, H\rangle |\omega_i, V\rangle + \mathcal{A}_{-\theta_{\text{deg}}}^{(2)}(\omega_s, \omega_i) S_-(z) |\omega_s, V\rangle |\omega_i, H\rangle \right] |z\rangle \\ & + \iiint d\omega_s d\omega_i dz \left[\mathcal{A}_{+\theta_{\text{deg}}}^{(2)}(\omega_s, \omega_i) S_+(z) |\omega_s, V\rangle |\omega_i, H\rangle + \mathcal{A}_{-\theta_{\text{deg}}}^{(1)}(\omega_s, \omega_i) S_-(z) |\omega_s, H\rangle |\omega_i, V\rangle \right] |z\rangle \quad (2) \end{aligned}$$

where S_\pm is the spatial distribution for each pumping beam along z . When nearly frequency degenerated photons are selected by filtering (*i.e.* $|\omega_s - \omega_i| < \sigma$), only the first integral is selected (see Fig. 2 in the main text). Finally the almost frequency indistinguishable two-photon state can be written as:

$$|\Psi(+\theta_{\text{deg}}, -\theta_{\text{deg}})\rangle = \iiint d\omega_s d\omega_i dz \left[\mathcal{A}_{+\theta_{\text{deg}}}^{(1)}(\omega_s, \omega_i) S_+(z) |\omega_s, H\rangle |\omega_i, V\rangle + \mathcal{A}_{-\theta_{\text{deg}}}^{(2)}(\omega_s, \omega_i) S_-(z) |\omega_s, V\rangle |\omega_i, H\rangle \right] |z\rangle \quad (3)$$

From now on, we simplify the notation, and write $\mathcal{A}_{+\theta_{\text{deg}}}^{(1)} \equiv \mathcal{A}_1$, $\mathcal{A}_{-\theta_{\text{deg}}}^{(2)} \equiv \mathcal{A}_2$, $S_+ \equiv S_1$ and $S_- \equiv S_2$.

1. Polarization only measurement

Since experimentally we perform polarization sensitive but frequency insensitive measurements, it is convenient to work with the reduced state taking the trace over frequencies and space coordinates in $|\Psi(+\theta_{\text{deg}}, -\theta_{\text{deg}})\rangle \langle \Psi(+\theta_{\text{deg}}, -\theta_{\text{deg}})|$:

$$\rho = \alpha_1 |HV\rangle \langle HV| + \alpha_2 |VH\rangle \langle VH| + \beta |VH\rangle \langle HV| + \beta^* |HV\rangle \langle VH| \quad (4)$$

with $\alpha_1 = \iiint d\omega_s d\omega_i dz |\mathcal{A}_1(\omega_s, \omega_i) S_1(z)|^2$, $\alpha_2 = \iiint d\omega_s d\omega_i dz |\mathcal{A}_2(\omega_s, \omega_i) S_2(z)|^2$ and $\beta = \iiint d\omega_s d\omega_i dz \mathcal{A}_1(\omega_s, \omega_i) \mathcal{A}_2^*(\omega_s, \omega_i) S_1(z) S_2^*(z)$

The reduced state can thus be represented as a 2×2 density matrix in the $(|HV\rangle, |VH\rangle)$ basis. The two eigenvalues λ_\pm are:

$$\begin{aligned} \lambda_\pm &= \frac{1}{2} \left[\alpha_1 + \alpha_2 \pm \sqrt{(\alpha_1 - \alpha_2)^2 + 4|\beta|^2} \right] \\ &= \frac{1}{2} \left\{ 1 \pm \sqrt{1 + 4 \left[|\beta|^2 - \alpha_1(1 - \alpha_1) \right]} \right\} \quad (5) \end{aligned}$$

where the normalization condition $\alpha_1 + \alpha_2 = 1$ has been used. Using this expression, all relevant quantities such as purity and entanglement can be computed since they depend on the integrals α_1 and $|\beta|^2$. The density matrix ρ is pure if and only if $\text{Tr}[\rho^2] = 1$, that is $\lambda_-^2 + \lambda_+^2 = 1$. Using $\lambda_- + \lambda_+ = 1$, this is equivalent to $\lambda_- = 0$, that is $|\beta|^2 = \alpha_1(1 - \alpha_1)$ (see Eq. (5)) (note that $|\beta|^2 \leq \frac{1}{4}$ since $\alpha_1 \leq 1$). In that case $\lambda_+ = 1$ and the corresponding eigenvectors are :

$$\lambda_+ = 1 \rightarrow \frac{1}{\sqrt{1 - \alpha_1}} \begin{pmatrix} \beta \\ 1 - \alpha_1 \end{pmatrix} \quad (6)$$

$$\lambda_- = 0 \rightarrow \frac{1}{\sqrt{\alpha_1}} \begin{pmatrix} \beta \\ -\alpha_1 \end{pmatrix} \quad (7)$$

ρ can thus be written as $\rho = |\phi\rangle\langle\phi|$ with:

$$|\phi\rangle = \frac{1}{\sqrt{1 - \alpha_1}} [\beta|HV\rangle + (1 - \alpha_1)|VH\rangle]. \quad (8)$$

The pure state is maximally entangled in polarization if the two, orthogonal, contributions have an equal weight, that is $|\beta|^2 = (1 - \alpha_1)^2$ *i.e.* when $\alpha_1 = \frac{1}{2} = \alpha_2$ and $|\beta|^2 = \frac{1}{4}$. These conditions can be arranged by optimizing the pumping conditions: to obtain a maximally entangled state in polarization (which is necessarily pure) we need to set up the two laser pumps such that there is perfect overlap between the joint spectral amplitudes of the generated pairs, as well as perfect spatial overlap between both pumping beams in the waveguide. The spectral overlap can be easily experimentally verified in our setup. However, ensuring the spatial overlap and perfect indistinguishability between the two different processes leading to photon pair generation is a more delicate task. This happens because the spatial transverse spatial profiles of the pumping beams are not the same, and thus cannot perfectly overlap. As a first approximation, we can consider that, after passing through the biprism, each pumping beam is a half gaussian. The point of maximum intensity of the two gaussians at the point they reach the sample is separated by a transverse (z) distance of δz . The spatial distribution of each beam can be modeled by the half-gaussian functions $S_1(z < 0) = 0$, $S_1(z > 0) = \sqrt{\frac{2\sqrt{2}}{w_p\pi}} e^{-\frac{z^2}{w_p^2}}$ and $S_2(z > \delta z) = 0$, $S_2(z < \delta z) = \sqrt{\frac{2\sqrt{2}}{w_p\pi}} e^{-\frac{(z - \delta z)^2}{w_p^2}}$. We see in Fig. 5 that, even in the case where the overlap between these two non-symmetric functions is maximized, we cannot reach $|\beta| = 0.5$.

2. Concurrence and linear entropy

We develop now our model to extract information about entanglement. Experimental measurements of the photon countings for each interaction indicate that condition $\alpha_1 = \alpha_2$ is satisfied. However, we can have $|\beta|^2 < 1/4$ and consequently non-maximally polarization entangled states if both beams are not in perfect frequency degeneracy and/or if their spatial profiles in the waveguide do not overlap. It was experimentally verified (see main text) that the degeneracy condition is satisfied within our spectral resolution. However, the overlap between the two pumping beams spatial profiles is more difficult to quantify, since it requires a perfect knowledge of the propagation effect on the laser beam intensity distribution throughout the optical path. Fig. 3 of the main text indicates the dependence of $|\beta|$ on the distance between the central points of the pumping beams in the z axis. The concurrence \mathcal{C} can be easily calculated, as a function of $|\beta|$, using its definition:

$$\mathcal{C} = \max\{0, \lambda_1 - \lambda_2 - \lambda_3 - \lambda_4\}, \quad (9)$$

where the λ_i are the ordered eigenvalues of the matrix R , defined as:

$$R = \sqrt{\rho \cdot \sigma_y \otimes \sigma_y \rho^T \sigma_y \otimes \sigma_y}. \quad (10)$$

Using the model introduced in the previous section, we have that $\mathcal{C} = 2|\beta|$ and thus the tangle $\mathcal{T} = 4|\beta|^2$. The linear entropy $S_L = 4/3(1 - \text{Tr}[\rho^2])$ can also be easily calculated in the conditions stated above. We have that $S_L = 2/3(1 - \mathcal{T})$ in this case. Notice that the value of $|\beta|$ quantities can be computed directly from a measurement of the experimental pumping beams profile. This model illustrates the important role played by the spatial overlap of the pumping beams and provides us with a clear guideline for controlling the degree of entanglement in polarization.

B. Frequency correlation control

Recent developments in quantum information theory have arisen a growing interest on generalized states of frequency correlation (like frequency-correlated, -anticorrelated or -uncorrelated photons). For all these reasons, several

techniques have been proposed to create two photon states with an arbitrary degree of entanglement and the counter-propagating phase-matching scheme has attracted a deal of attention because of its unusual flexibility in the control of the quantum properties of the emitted photons. Frequency correlation is governed by the joint spectral amplitude (JSA), the probability amplitude for an idler photon to have a frequency ω_i when its signal twin has a frequency ω_s . It consists of the product of the pump beam spectral amplitude and the phase-matching distribution special to the nonlinear waveguide used, and it can be expressed as follows:

$$\text{JSA}(\omega_s, \omega_i) = A \alpha(\omega_s + \omega_i) \Phi(\omega_s, \omega_i, \theta) \quad (11)$$

where A is a normalization factor, α is the spectral amplitude of the pump beam, and Φ is the phase-matching function depending on the pump incident angle θ . As a pump distribution of the mode-locked picosecond Ti:Sa laser, we assume a spectrum with sech shape. Taking into account the transmission function of the integrated microcavity of the waveguide, we arrive at

$$\alpha(\omega) \propto \text{sech}\left(1.7628 \frac{\omega_p - \omega}{\sigma_p}\right) \times \frac{1}{1 - 4 \frac{\omega_p - \omega}{\sigma_{\text{cav}}}} \quad (12)$$

with ω_p the pump central frequency, σ_p the spectral pump width, σ_{cav} the cavity resonance width. The cavity resonance frequency we assume to be coincident with the pump central frequency ω_p .

In general, the form of the phasematching function Φ depends only on the phase mismatch Δk and is determined by the spatial pump amplitude function $g(z)$ in waveguide direction:

$$\Phi(\Delta k) = \int dz g(z) e^{-2i\pi\Delta k z} \quad (13)$$

For a standard co-propagating phasematching scheme in a waveguide, Φ will take the form of the sinc function. In our transverse pump configuration we have to take into account the spatial pump profile. If we assume this as a Gaussian e^{-z^2/w_p^2} , we find in good approximation for a waveguide placed at the beam's waist

$$\Phi(\Delta k) \propto e^{-\frac{1}{4}w_p^2\Delta k^2}. \quad (14)$$

For an incidence angle θ , the phase-mismatch for interaction 1 is given by

$$\begin{aligned} \Delta k &= \frac{\sin\theta(\omega_s + \omega_i)}{c} - \frac{n_H\omega_s}{c} + \frac{n_V\omega_i}{c} \\ &= \frac{1}{c} [(\sin\theta - n_H)\omega_s + (\sin\theta - n_V)\omega_i] \end{aligned} \quad (15)$$

Thus, we can write as JSA function:

$$\text{JSA}(\omega_s, \omega_i) = A \text{sech}\left(1.7628 \frac{\omega_p - \omega_s - \omega_i}{\sigma_p}\right) \frac{1}{1 - 4 \frac{\omega_p - \omega_s - \omega_i}{\sigma_{\text{cav}}}} e^{-\frac{w_p^2}{4c^2} [(\sin\theta - n_H)\omega_s + (\sin\theta - n_V)\omega_i]^2} \quad (16)$$

In order to quantify the degree of correlation of a given frequency state and its eventual separability, one has to perform a Schmidt decomposition of the JSA, a basis transformation into a set of orthogonal Schmidt modes $\{\psi_n; \varphi_n\}$:

$$\text{JSA}(\omega_s, \omega_i) = \sum_n \sqrt{\lambda_n} \psi_n(\omega_s) \varphi_n(\omega_i) \quad (17)$$

with the normalization condition $\sum_n \lambda_n = 1$.

If the sum of expression (17) contains only 1 term, the JSA is factorizable and the photons are uncorrelated in frequency. On the contrary, if it contains a great number N of terms, the frequency state is strongly correlated (or anti-correlated).

In Figure 1, we present three examples of calculated JSA shapes and their corresponding Schmidt decompositions for interaction 1 at degeneracy. These states have been calculated with our source for three different pumping configurations in the picosecond regime. We note that the versatility in the tuning of the frequency state is peculiar to the counterpropagating geometry, since in this case the pump bandwidth σ_p and the sample length L each act on the length of one of the ellipse axes ($\omega_s + \omega_i = \omega_p^0$ and $\omega_s - \omega_i = 0$). Thus, by varying independently these two parameters, every possible eccentricity of the ellipse can be obtained. In the case of copropagating geometry instead, in most cases, only one of the lengths of the axis of the ellipse can be adjusted, which usually prevents the obtention of other states than frequency-anti-correlated ones. More exotic frequency states presenting JSA with several lobes can be obtained by playing also with the phase of the pump beam; an experimental and theoretical work on this kind of states is in progress.

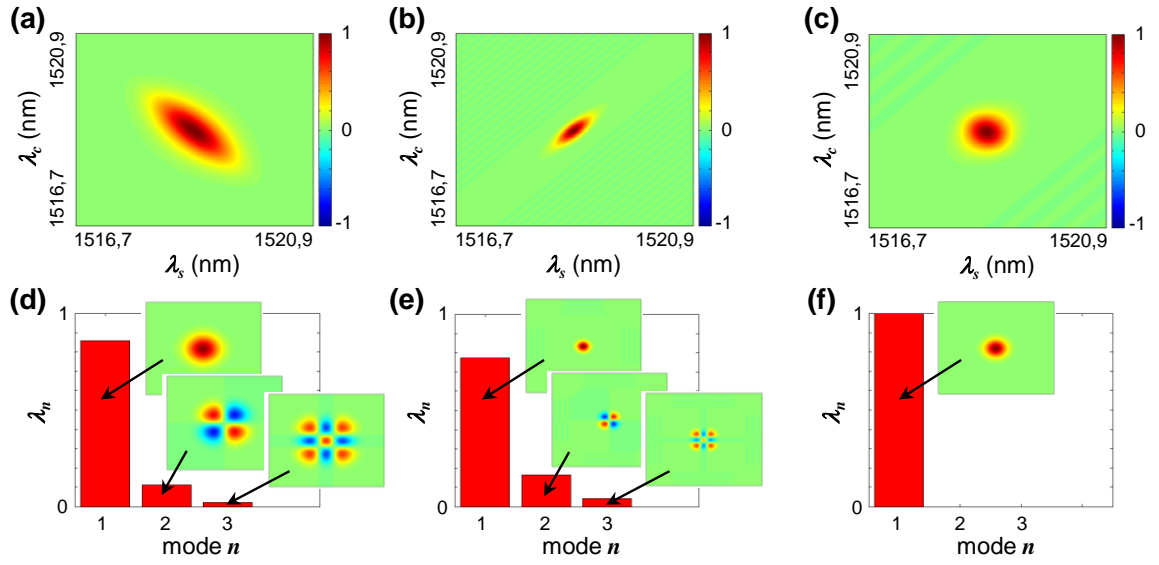


FIG. 1: Calculated JSA for our AlGaAs waveguide device with a pump having a pulse duration 3.5 ps for different pump beam waists and their respective Schmidt decompositions. All three cases of negative correlation (pump waist = 0.2 mm) (a), positive correlation (pump waist = 1.3 mm) (b) and decorrelation (pump waist = 0.45 mm) (c) can be achieved. Accordingly, the Schmidt decomposition (f) of case (c) has only one Schmidt mode pair with $\lambda_0 = 1$.