Supplementary Materials for

Spatially entangled photon pairs from lithium niobate nonlocal metasurfaces

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S1. COUPLED-MODE THEORY OF NONLOCAL METASURFACES

In our work, we consider a dielectric grating which induces high-Q guided-mode resonances (GMRs) inside the lithium niobate (LN) slab and accordingly enhances the nonlinear processes. We note that the nonlinear wave mixing mediated by GMRs can have different features from surface lattice resonances (SLR) (22, 23, 48, 49) which arise from the collective excitation of plasmonic particle resonances that couples the propagating Rayleigh anomalies with localized surface plasmons (50).

As shown in the main paper, the SiO$_2$ grating on lithium niobate structure supports two resonances when the input polarization is along the grating. These two modes couple with each other and the coupling strength is linearly proportional to the transverse wavenumber across the grating ($k_y$). In addition, we also find that the eigenfrequencies show a weak quadratic dependence on the transverse wavenumber along the grating ($k_z$). In the following, we label these two modes as mode 1 and mode 2, which have resonance frequencies at $\omega_1 + i\gamma_1$ and $\omega_2 + i\gamma_2$, respectively, at normal incidence. The imaginary parts of the eigenfrequencies indicate the loss of the modes, which determine the quality factors of each mode through $Q_n = \omega_n / (2\gamma_n)$. In our structure the mode loss comes from the radiation loss caused by the grating. Figure S1 shows the field profiles ($E_z$ component) of mode 1 and 2 for a zero transverse wavenumber. As can be seen, modes 1 and 2 are anti-symmetrical and symmetrical, respectively, along the $y$ direction. The anti-symmetrical mode 1 cannot be excited by or radiate into plan waves in free space, thus it has infinite quality factor. It is
called a dark mode or bound state in the continuum.

The metasurface also supports other two resonances when the input polarization is perpendicular to the grating. The variations of their inter-coupling and eigenfrequencies relative to the transverse wavenumber are analogous to modes 1 and 2. We label these two modes as 3 and 4, which have complex resonance frequencies at $\omega_3 + i\gamma_3$ and $\omega_4 + i\gamma_4$, respectively, at normal incidence. Modes 3/4 can also couple with the modes 1/2 for non-zero $k_z$. Specifically, mode 3 only couples with mode 1 and mode 4 only couples with mode 2, due to their symmetry properties. The coupling strength is linearly proportional to $k_z$.

Based on the above considerations, we formulate the temporal coupled mode theory following the general methodology (38) under the conditions of no material absorption,

$$\frac{da}{dt} = (i\Lambda - \Gamma)a + \kappa^T s_i,$$

$$s_o = S s_i + \kappa a.$$  \hspace{1cm} (S1, S2)

Here $a$ are the amplitudes of four modes in the metasurface,

$$a = \begin{pmatrix} a_1 \\ a_2 \\ a_3 \\ a_4 \end{pmatrix}.$$ \hspace{1cm} (S3)

Figure S1. **Mode profiles.** Normalized $E_z$ distributions of (a) mode 1 and (b) mode 2 for zero transverse wavenumber, i.e. $k_y = k_z = 0$. Note that the $E_x$ and $E_y$ components are zero for these two modes.
and $s_{i,o}$ are the amplitudes of the incident (i) and outgoing (o) plane waves, defined as

$$s_{i/o} = \begin{pmatrix} s_{i/o,z^+} \\ s_{i/o,z^-} \\ s_{i/o,y^+} \\ s_{i/o,y^-} \end{pmatrix},$$

(S4)

where $+/-$ indicate the regions above (+) or below (-) the metasurface, and $z/y$ denote the
dominant direction of wave polarization.

The coupling matrix can be written as

$$\Lambda = \begin{bmatrix} \omega_1 + U_{12,z}k_x^2 + U_{12,y}k_y^2 & iV_{12}k_y & -iC_{13}k_z & 0 \\ -iV_{12}^*k_y & \omega_2 + U_{12,z}k_x^2 + U_{12,y}k_y^2 & 0 & -iC_{24}k_z \\ iC_{13}^*k_z & 0 & \omega_3 + U_{34,z}k_x^2 + U_{34,y}k_y^2 & iV_{34}k_y \\ 0 & iC_{24}^*k_z & -iV_{34}^*k_y & \omega_4 + U_{34,z}k_x^2 + U_{34,y}k_y^2 \end{bmatrix},$$

(S5)

and the losses arise through the radiation channels as

$$\Gamma = \begin{bmatrix} \gamma_1 & 0 & 0 & 0 \\ 0 & \gamma_2 & 0 & 0 \\ 0 & 0 & \gamma_3 & 0 \\ 0 & 0 & 0 & \gamma_4 \end{bmatrix}.$$

(S6)

Due to the structure symmetry, modes 1 and 4 are bound states in continuum (BIC) that
do not radiate, such that $\gamma_1 = \gamma_4 = 0$. Then, the excitation matrix has the form

$$\kappa^T = \begin{bmatrix} 0 & 0 & 0 & 0 \\ \kappa_{2+} & \kappa_{2-} & 0 & 0 \\ 0 & 0 & \kappa_{3+} & \kappa_{3-} \\ 0 & 0 & 0 & 0 \end{bmatrix}.$$

(S7)

According to the general coupled-mode theory (38), $\kappa^T \kappa = 2\Gamma$, which for our system implies

$$|\kappa_{2-}|^2 + |\kappa_{2+}|^2 = 2\gamma_2,$$

$$|\kappa_{3-}|^2 + |\kappa_{3+}|^2 = 2\gamma_3.$$

(S8)

We note that the $S$ matrix is unitary and symmetric, and also $SK^* = -K$, such that its
form is

\[ S = \begin{bmatrix} r_{2+} & t_2 & 0 & 0 \\ t_2 & r_{2-} & 0 & 0 \\ 0 & 0 & r_{3+} & t_3 \\ 0 & 0 & t_3 & r_{3-} \end{bmatrix} \]  
(S9)

where \( |r_{2+}|^2 = |r_{2-}|^2 = 1 - |t_2|^2 \) and \( |r_{3+}|^2 = |r_{3-}|^2 = 1 - |t_3|^2 \).

If only one wave with the frequency \( \omega \) is incident on the metasurface, for example \( s_{i,z+} = \exp(i\omega t) \) and other input amplitudes are zero, then

\[ a = (i\omega - i\Lambda + \Gamma)^{-1} \begin{pmatrix} 0 \\ \kappa_{2+} \\ 0 \\ 0 \end{pmatrix} \exp(i\omega t). \]  
(S10)

It is convenient to perform eigenmode decomposition as

\[ A = W^{-1} a, \]  
(S11)

\[ \Lambda + i\Gamma = W\Omega W^{-1}, \]  
(S12)

where \( \Omega \) is a diagonal matrix with eigenvalues, and \( W \) is a matrix with corresponding eigenvectors arranged in columns. Then, we use Eq. (S10) to find the amplitudes of excited modes as

\[ i\omega A = i\Omega A + W^{-1}\kappa^T s_i \exp(i\omega t), \]  
(S13)

or equivalently

\[ A_m = -i(\omega - \Omega_m)^{-1}\tilde{A}_m \exp(i\omega t), \]  
(S14)

where \( m \) is the mode number and

\[ \tilde{A}_m = [W^{-1}\kappa^T s_i]_m. \]  
(S15)

For our experimental structure, we obtain the values of each parameter through numerical simulations and fittings. For example, \( \omega_{1,2,3,4} \) and \( \gamma_{1,2,3,4} \) are directly obtained by calculating the eigenfrequencies \( \Omega \) at normal incidence. Then, by letting \( k_y = 0 \) and calculating the four eigenfrequencies at different \( k_z \), we can obtain the values of \( V_{12} \) and \( V_{34} \) by numerically fitting the numerical results. Similarly, the values of \( U_{12,34} \) and \( C_{13,24} \) are obtained by numerically fitting the eigenfrequencies at different \( k_z \) when \( k_y \) is set to zero. Table S1 lists the values of
Table S1. **Coupled-mode theory coefficients.** The values are determined through finite-element simulations for the fabricated meta-grating geometry.

| Eigenfrequencies at normal incidence, THz |
|------------------------------------------|
| $\omega_1/(2\pi)$ | $\omega_2/(2\pi)$ | $\omega_3/(2\pi)$ | $\omega_4/(2\pi)$ |
| 190.21 | 190.97 | 206.89 | 207.95 |

| Loss coefficients at normal incidence, THz |
|------------------------------------------|
| $\gamma_1/(2\pi)$ | $\gamma_2/(2\pi)$ | $\gamma_3/(2\pi)$ | $\gamma_4/(2\pi)$ |
| 0 | 0.18 | 0.25 | 0 |

| Coupling coefficients, THz·μm |
|-----------------------------|
| $V_{12}/(2\pi)$ | $V_{34}/(2\pi)$ | $C_{13}/(2\pi)$ | $C_{24}/(2\pi)$ |
| 22.61 | 22.40 | 2.07 | 2.20 |

| Coupling coefficients, THz·μm² |
|-----------------------------|
| $U_{12,y}/(2\pi)$ | $U_{34,y}/(2\pi)$ | $U_{12,z}/(2\pi)$ | $U_{34,z}/(2\pi)$ |
| -0.18 | -0.64 | 1.78 | 1.33 |

| Nonlinear coefficients, ($\mu$m²/mW)¹/² |
|---------------------------------------|
| $\eta_1$ | $\eta_2$ | $\eta_3$ | $\eta_4$ |
| $1.98 \times 10^{-5}$ | $1.98 \times 10^{-5}$ | 0 | 0 |

all the CMT coefficients for the metasurface with the geometrical parameters $a = 886$ nm, $w = 443$ nm, $h = 200$ nm, and $t = 304.3$ nm.

Figures S2a-d show the frequencies and quality factors of modes 1 and 2 predicted by the CMT and the finite-element simulations in two cases, $k_z = 0$ and $k_y = 0$. The CMT and simulated results are very similar. Based on the CMT, we also calculate the eigenfrequency and thus phase/energy matching map as a function of the transverse wavenumber, as shown in Fig. S2f. The result is almost identical to the one obtained by direct finite-element modelling, c.f. Fig. S2e. These comparisons confirm that the CMT can accurately describe the optical resonances of the metasurface at the photon-pair wavelengths.

We note that a higher-order guided-mode resonance closest to the operating pump wavelength (around 785 nm) is strongly detuned at $\sim 710$ nm, see Fig. S3a. Therefore, we do not take into account the guided-mode resonance effect at the pump wavelength in the coupled-mode theory and data analysis.
where due to energy and momentum conservation, \( \omega^{(p)} = \omega^{(s)} + \omega^{(i)} \) and \( k^{(p)} = k^{(s)} + k^{(i)} \), see Fig. S3b. Here the superscripts refer to the signal (s), idler (i), and pump (p) waves, and \( k = (k_y, k_z) \) are the transverse wavevectors in air. The coefficients \( \eta_n \) are proportional to the nonlinear mode overlaps and the quadratic susceptibility, and they are approximately constant across the spectral and spatial bandwidths of photons. In our structure, the dominant coefficients are \( \eta_1 \approx \eta_2 \), whereas \( \eta_3 \approx \eta_4 \approx 0 \).

We use Eqs. (S11) and (S15) to obtain

\[
\Xi^{(s)}(\omega^{(s)}, \omega^{(i)}, k^{(s)}, k^{(i)}) = \frac{1}{I^{(s)} I^{(i)} 4(2\pi)^3} \left| \sum_{m_s, m_i} \tilde{W}_{m_s, m_i} A_{m_s}^{(s)}(\omega^{(s)}, k^{(s)}) A_{m_i}^{(i)}(\omega^{(i)}, k^{(i)}) \right|^2 \tag{S17}
\]

where \( \tilde{W}_{m_s, m_i} = \sum_n \eta_n W_{n, m_s}^{(s)} W_{n, m_i}^{(i)} \).

According to the classical-quantum correspondence \((17, 19, 51)\), the photon-pair rate for a spectral region close to degeneracy is proportional to

\[
\Xi(\omega^{(s)}, \omega^{(i)}, k^{(s)}, k^{(i)}) = \frac{1}{I^{(s)} I^{(i)} 4(2\pi)^3} \left| \sum_{m_s, m_i} \tilde{W}_{m_s, m_i} A_{m_s}^{(s)}(\omega^{(s)}, k^{(s)}) A_{m_i}^{(i)}(\omega^{(i)}, k^{(i)}) \right|^2 \tag{S18}
\]

where \( I^{(s)} \) and \( I^{(i)} \) are the incident signal and idler intensities in the SFG calculation of \( b \).

The emission integrated over all frequencies is

\[
\Xi_k(k^{(s)}, k^{(i)}) = \int d\omega^{(s)} \Xi(\omega^{(s)}, \omega^{(i)} = \omega^{(p)} - \omega^{(s)}, k^{(s)}, k^{(i)})
\]

\[
= \frac{1}{I^{(s)} I^{(i)} 4(2\pi)^3} \int d\omega^{(s)} \left| \sum_{m_s, m_i} \tilde{W}_{m_s, m_i} A_{m_s}^{(s)}(\omega^{(s)}, k^{(s)}) A_{m_i}^{(i)}(\omega^{(i)} = \omega^{(p)} - \omega^{(s)}, k^{(i)}) \right|^2
\]

\[
= \frac{-2\pi i}{I^{(s)} I^{(i)} 4(2\pi)^3} \sum_{m_{s1}, m_{s1}, m_{s2}, m_{i2}} \tilde{W}_{m_{s1}, m_{i1}} \tilde{W}_{m_{s2}, m_{i2}} A_{m_{s1}}^{(s)} A_{m_{i1}}^{(i)} A_{m_{s2}}^{(s)*} A_{m_{i2}}^{(i)*}
\times (\Omega^{(s)}_{m_{s1}} + \Omega^{(i)}_{m_{i1}} - \Omega^{(s)*}_{m_{s2}} - \Omega^{(i)*}_{m_{i2}})
\times (\Omega^{(s)}_{m_{s1}} - \Omega^{(s)*}_{m_{s2}})^{-1} (\Omega^{(i)}_{m_{i1}} - \Omega^{(i)*}_{m_{i2}})^{-1}
\times (\Omega^{(s)}_{m_{i1}} + \Omega^{(i)}_{m_{i1}} - \omega^{(p)})^{-1} (-\Omega^{(s)*}_{m_{s2}} - \Omega^{(i)*}_{m_{i2}} + \omega^{(p)})^{-1} \tag{S19}
\]
We use the CMT to calculate the photon-pair rates in the main manuscript, and also present the supplementary plots in Figs. S3c, d and S4. Interestingly, Fig. S3d shows that the SPDC rate for a plane-wave pump at $\lambda_p = 1573/2$ nm, which double wavelength is in the gap between the upper and lower branches of the signal and idler modes, features a peak at a non-zero wavevector $\sim 0.015 \text{ rad}/\mu\text{m}$. This explains why a Gaussian pump has a larger SPDC rate than a normally incident plane-wave pump in the wavelength range around 1573 nm, as observed in Fig. 1f and Fig. S3c.
Figure S2. **Mode dispersion.** a-d Frequencies and quality factors of modes 1 and 2 calculated by finite-difference modelling and predicted by the CMT as a function of (a,b) $k_y$ when $k_z = 0$ and (c,d) $k_z$ when $k_y = 0$. e,f Frequencies of mode 2 (e) calculated by finite-difference modelling and (f) predicted by the CMT as functions of $k_y$ and $k_z$. The black dashed contour lines mark $\omega_2 = 191.19$ THz, which is the same phase matching line as in Fig. 1d of the main paper, but shown here in a extended $k_z$ and narrower $k_y$ region.
Figure S3. **Phase matching and simulated SPDC rates.**

- **a** Simulated transmittance and reflectance of the metasurface at normal incidence around the pump wavelength range.
- **b** Diagram of the transverse phase matching of the SPDC in the free-space far field and in the near field inside the LiNbO$_3$ thin film, in the regime of guided mode resonances at the signal and idler wavelengths (the labels of signal and idler photons can be interchanged).
- **c** CMT predicted SPDC rate as a function of the pump wavelength when the collection angle is $0.7^\circ$, which corresponds to the collection angle in the experiment. Results are presented for a pump in the form of a plane-wave (solid line) and a Gaussian beam with $100\mu m$ diameter (dashed line). For comparison, note that the simulations in Figs. 1d, 1e, and 1f of the main manuscript are performed for a broader collection angle of $3.2^\circ$.
- **d** Transverse wavevector distribution of the Gaussian pump at $\lambda_p = 1573/2$ nm with a beam diameter of $100\mu m$ (dotted line) and the simulated SPDC rate for plane-wave pumps at different transverse wavevectors along the $y$ direction (solid line).
Figure S4. **Simulated enhancement of SPDC.** a,b Simulated enhancement of (a) the peak SPDC spectral brightness and (b) peak SPDC total rate compared to an unpatterned film as a function of the collection angle range. In (b), the photon bandwidth of 50 nm around the 1570 nm wavelength is considered, corresponding to the bandpass filter in the experimental setup. Note that the collection angle in the experiment is $\sim 0.7^\circ$. The enhancement reduces for larger collection angles because the metasurface emits in a particular angular range according to the transverse phase-matching, whereas emission from an unpatterned thin-film is almost constant across a broad range of angles.
S2. EXPERIMENTAL SETUPS: IMPLEMENTATION AND CALIBRATION

S2.1. Full experimental setup for quantum measurements

The full setup for quantum experiments is shown in Fig. S5. A laser beam with a wavelength at 785 nm is focused on a metasurface fabricated on top of a lithium niobate film to produce photon pairs. The photon emission is collimated using a lens with a diameter of one inch. Both focusing lens and collimating lens have a focal length of 100 mm. The photon pairs are collected with a fiber whose numerical aperture is 0.27. The aperture diameter of the fiber collimator is 5 mm, suggesting a collection angle of \( \sim 0.7^\circ \). The collected photon pairs pass through a 50:50 fiber beam splitter, and their coincidence is then registered by two single-photon detectors. Several spectral filters are introduced to remove the fluorescence produced by the optics and metasurface, including a shortpass at

![Experimental setup diagram](image-url)

Figure S5. **Experimental setup.** The coincidence measurement is performed with a Hanbury Brown-Twiss setup. The spatial correlations of generated photons pairs are characterized using an aperture whose size is larger than the beam profile of the collected photon pairs.
850 nm, a long pass at 1100 nm and a bandpass at 1570 nm with a 50 nm FWHM. A half waveplate placed in front of the focusing lens is used to control the pump polarization. We employ a polarizer after the photon emission to analyze the polarization of photon pairs.

We experimentally characterize the spatial correlations of photon pairs by transversely translating an aperture placed after the photon emission.

S2.2. Linear transmission spectrum of the metasurface

The linear transmission of the nonlocal metasurface is measured with a tunable CW laser and a broadband thermal lamp. Either light source has its limitations, as can be observed in Figs. S6b,c. The CW laser has a short wavelength tuning range up to 1575 nm. As a result, the mode splitting at a large incident angle cannot be observed with this laser. The tungsten-halogen lamp enables measurements over an ultra-broad bandwidth. Because of the limited spectrometer resolution of 2.4 nm, however, measurements with the lamp cannot capture the fine structure of the resonance and do not resolve the minimum of the transmission dip.

Figure S6. Linear transmission measurements of the metasurface. a top view of the SEM image of the fabricated metasurface. b Transmission measurements with a tunable CW laser. The grey curve plots the experimental data and the red curve presents the corresponding fitting results for a Fano resonance. c Transmission measurements with a white lamp. The blue curve is for normal incidence and the yellow curve is for an incidence angle of 1°.
Figure S7. **Pump laser wavelength vs. temperature.** The grey shaded regions indicate mode hopping. This experimental characterization data is used to measure the wavelength dependence shown in Fig. 3c of the main paper.

In Fig. S6b, there are several small peaks with the same differences in wavelengths. This is due to the Fabry-Perot effect introduced by the substrate. We perform a nonlinear fitting (red curve) of the measured transmission with a Fano resonance function and extract the value of Q factor and resonant wavelength as $\sim 455$ and 1570.5 nm, respectively. In Fig. S6c, we show the metasurface transmission measured with the broadband lamp source at normal incidence and an incident angle of 1°. An apparent mode splitting is identified at off-normal incidence.

**S2.3. Characterization of pump laser for SPDC**

The wavelength of the pump laser can be tuned by the diode temperature, as shown in Fig. S7. We find that the wavelength is not linearly dependent on the temperature. Additionally, mode hopping of the laser diode is present at certain temperatures. We avoid the wavelengths next to the hopping regions when measuring the photon-pair coincidences. As a result, the wavelength dependence discussed in Fig. 3c of the main paper shows scattered data points.

The choice of the beam diameter is important for the efficient excitation of grating res-
Figure S8. **Experimentally measured second-order correlations.** Characteristic examples of $g^{(2)}(\tau)$ of the photon pairs, emitted from a metasurface and b unpatterned structure.

In our work, we selected the pump beam diameter of 100 $\mu$m based on the following considerations. First, to efficiently excite the guided-mode resonances of the grating, the beam diameter should be larger than the transverse propagation distance of the signal or idler modes, which is estimated to be 71 $\mu$m in the main manuscript. Second, the beam diameter has to be smaller than our metasurface size of 400 $\mu$m.

**S2.4. Second-order correlations of produced photon pairs**

The normalized second-order correlation $g^{(2)}(\tau)$ of the produced photon pairs at time delay $\tau$ can be obtained from (54)

$$g^{(2)}(\tau) = \frac{N_c(\tau)}{N_1N_2T_c},$$

where $N_c(\tau)$ is the total coincidence rate at delay $\tau$, $T_c$ is the time window of registering coincidence, and $N_1$ and $N_2$ are the single photon count rates collected by the two detectors. The coincidence to accidental ratio (CAR) can be calculated as $\text{CAR} = g^{(2)}(0) - 1$. The $g^{(2)}(\tau)$ of photon pairs from metasurface and unpatterned film are calculated using Eq. (S20), and characteristic results are shown in Fig. S8. The value of $g^{(2)}(0)$ calibrated from the
metasurface resonantly-enhanced emission is \( \sim 5000 \), while \( g^{(2)}(0) \) is only \( \sim 13 \) for photon pairs emitted from an unpatterned film. In both cases, the values of CAR exceed the classical bound of 2.

We estimate the overall detection efficiency for the photon pairs from the efficiencies of collection and detectors. The efficiency of single-photon detectors calibrated by the manufacturer is \( \eta_{\text{det}} = 25\% \). The collection efficiency for each photon is estimated as \( \eta_{\text{col}} = 25\% \sim 30\% \), taking into account the losses from the collection lens, reflecting mirrors, filters and the fiber coupling. The overall efficiency \( \eta_{\text{tot}} \) for detecting both photons can then be calculated as

\[
\eta_{\text{tot}} = (\eta_{\text{det}}\eta_{\text{col}})^2, \tag{S21}
\]

which gives an estimate from 0.4% to 0.6%. 
In this section, we formulate the Cauchy-Schwartz inequality (CSI) and discuss its violation for two-photon correlations under our experimental conditions, which serves as a witness of multi-mode spatial entanglement.

Let us consider the spatial correlations between two complementary spatial regions, \( A_1 \) and \( A_2 \). As shown in Fig. S9a, for experimental measurement with an aperture scanned along the \( q \) (\( q = y, z \)) direction (the aperture size \( L \) is much larger than the beam size \( d_q \) of the emitted or collected photons), we define \( A_1 \) as \( q \leq q_s \) and \( A_2 \) as \( q \geq q_s \). Here, \( q = 0 \) is the centre position of the beam.

We define \( \Gamma_{n,m} \) \((n, m = 1, 2)\) as the spatial correlations between regions \( A_n \) and \( A_m \). For classical light, the correlations can be found through the averaging over instantaneous intensities as,

\[
\Gamma_{n,m} = \langle I_n I_m \rangle = \int I_n(t)I_m(t) dt ,
\]  

(S22)

where \( I_n(t) = \int_{A_n} I(t, r) dr \).

We use Cauchy–Schwarz inequality to find that

\[
\Gamma_{1,2} \leq \sqrt{\Gamma_{1,1} \Gamma_{2,2}} .
\]  

(S23)

Figure S9. **Diagram of aperture positions.** a The center of the aperture is aligned with the center of the beam profile, defining the zero position of the aperture. Two spatial regions \( A_1 \) and \( A_2 \) are separated at the position \( q_s \). b-c The aperture is moved to the position (b) \( q = q_s - L/2 \) or (c) \( q = q_s + L/2 \). The coincidence measurements of the transmitted photons indicate the auto-correlation for the region (b) \( A_1 \) or (c) \( A_2 \).
Let us denote as $\Gamma_t$ the correlations over the whole region, including $A_1$ and $A_2$,

$$\Gamma_t = \langle (I_1 + I_2)^2 \rangle = \Gamma_{1,1} + \Gamma_{2,2} + 2\Gamma_{1,2} \quad (S24)$$

Then, using Eqs. (S23) and (S24), we obtain

$$\Gamma_t - \Gamma_{1,1} - \Gamma_{2,2} = 2\Gamma_{1,2} \leq 2\sqrt{\Gamma_{1,1}\Gamma_{2,2}}, \quad (S25)$$

and finally

$$\left(\sqrt{\Gamma_{1,1}} + \sqrt{\Gamma_{2,2}}\right)^2 \geq \Gamma_t. \quad (S26)$$

Violation of the equation above indicates non-classical correlations and witnesses the presence of multi-mode spatial entanglement (46, 55). This effect is known in the literature as the violation of classical Cauchy-Schwartz inequality (56, 57). In our case, it is due to a spatial analogue of photon antibunching (47, 58). Interestingly, similar to Eq. (5) in Ref. (59), the CSI in Eq. (S26) involves the correlation functions integrated over momenta, since the aperture is located in the diffraction far-field where the transverse positions correspond to momenta of the photons with which they are emitted from the metasurface.

For the beam size $d$, we need to consider the range of aperture positions

$$-\frac{d}{2} \leq q_s \leq \frac{d}{2}. \quad (S27)$$

The value of $d$ can be estimated based on the width of a 'flat' region where $C(q) \simeq C(0)$ for $|q| \leq (L - d)/2$.

For the the analysis of photon-pair measurements, we express the CSI given in Eq. (S26) in terms of the two-photon coincidences $C(q)$ as a function of aperture position $q$ ($q = y$ or $q = z$) can be measured experimentally, as shown in Figs. 4c-d of the main paper. In terms of Figs. 9b-c, the auto correlations of regions $A_1$ and $A_2$ are $\Gamma_{1,1} = C(q_s - L/2)$ and $\Gamma_{2,2} = C(q_s + L/2)$, respectively. The total correlations, when the beam is not blocked, corresponding to the aperture position in the middle, are $\Gamma_t = C(0)$. We can therefore rewrite Eq. (S26) as follows

$$\Gamma(q_s) \equiv \left(\sqrt{C(q_s - L/2)} + \sqrt{C(q_s + L/2)}\right)^2 \geq \Gamma_t = C(0). \quad (S28)$$

Note that in the symmetric case when half of the beam is blocked, Eq. (S28) reduces to

$$4\Gamma_{1,1} = 4\Gamma_{2,2} = 4C(\pm L/2) \geq \Gamma_t = C(0).$$
Figure S10. **Calculated coincidence vs. aperture position.** Normalised simulated coincidences $C$ as a function of the aperture position $q$ along the $a,c$ $y$ and $b,d$ $z$ directions for the collection angles of $a,b$ 0.7 degrees, matching the experimental conditions, and $c,d$ 1.4 degrees, twice the experimental range. The aperture size $L$ is set to 1.3 times of the collected beam size at both collection angles. A Gaussian pump with a diameter of 100 $\mu m$ is considered in the simulation.

We present the simulated dependence of the coincidence $C(q)$ vs. aperture position along the $y$ or $z$ axis, for two different collection angles, in Fig. S10. We find a good agreement between the experimental results in Figs. 4$c,d$ and the theoretical modelling for the same collection angle in Figs. S10$a,b$. The simulations predict that as the collection angle is increased, the coincidence curves remain almost the same for the aperture translation along the z direction [Figs. S10$b$ and d], which happens because the emission pattern in momentum space is nearly flat along this axis as shown in Fig. 1$d$. On the other hand, for a larger collection angle the dependence along the $y$ direction is visibly modified and its difference from the $z$ direction becomes more apparent, c.f. Figs. S10$c$ and d, which reflects the asymmetry of the emission pattern.
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