Electronic Supplementary Material (ESI) for Nanoscale.

Photon pairs bi-directionally emitted from a resonant metasurface: supplementary information

CHANGJIN SON[1,2], **VITALIY SULTANOV**[1,2], **TOMÁS SANTIAGO-CRUZ**[1,2], **ARAVIND P. ANTHUR**[3], **HAIZHONG ZHANG**[3], **RAMON PANIAGUA-DOMINGUEZ**[3], **LEONID KRIVITSKY**[3], **ARSENIY I. KUZNETSOV**[3], AND **MARIA V. CHEKHOVA**[1,2]

[1] *Max Planck Institute for the Science of Light, 91058 Erlangen, Germany. E-mail: changjin.son@mpl.mpg.de*
[2] *Friedrich-Alexander Universität Erlangen-Nürnberg, 91058 Erlangen, Germany*
[3] *A*STAR (Agency for Science, Technology and Research) Research Entities, Institute of Materials Research and Engineering, Singapore 138634 Singapore*

1. METHODS

Polarization properties of the second harmonic generation

The nonlinear response of the metasurface (MS) is characterized through the sum-frequency generation (SHG). The setup is shown in Fig. S2. The beam from an optical parametric amplifier (OPA, 1 kHz repetition rate, 20 ps pulse duration) is used as a pump [1]. The pump beam is horizontally polarized with polarization beamsplitter PBS. To monitor the pump beam, a 70:30 (transmission:reflection) beamsplitter (BS) is mounted. The reflected beam is sent to an infrared spectrometer. The transmitted beam passes through half-wave plate HWP1 and is focused on the metasurface by a C-coated achromatic lens with the 50 mm focal distance. The generated SHG is collected by another lens with the same focal distance. The pump beam is filtered out by using a short-pass filter with the cut-off wavelength 950 nm. The polarization of the second harmonic radiation is analyzed by an analyzer, which is another half-wave plate (HWP2) and a B-coated Glan-Thomson prism (GT). Finally, the signal after the analyzer is sent to a visible spectrometer. First, we measure the SHG efficiency as a function of the pump polarization. After finding the optimal polarization of the pump (horizontal, along the net dipole moment of the resonant mode), we fix it and measure the polarization of the second-harmonic radiation.

Polarization dependence of spontaneous parametric down-conversion

We measured the dependence of spontaneous parametric down-conversion (SPDC) rate on the pump polarization for three measurement geometries (forward-forward, forward-backward, and backward-backward). To control the pump polarization, a half-wave plate is mounted right after the pump laser in the correlation measurement experimental setup. The measured dependence is shown in Fig. S3(a-c) for the forward-forward, forward-backward, and backward-backward configurations, respectively. Note that in all three configurations the data exhibit the same dependence observed when a polarization analyzer is rotated in the SHG experiments, see Fig. S2(c).

White light measurement

To measure the transmission spectrum of the MS, we use supercontinuum radiation (1 kHz repetition rate, 20 ps pulse duration) with the polarization set as the resonance polarization of the MS (horizontal). The beam, initially 1.5 mm in diameter, is focused on the MS by a C-coated achromatic lens with the 100 mm focal length. The spot size at the focus is $\sim 100\ \mu$m at 1200 nm. The transmitted light from the MS is collimated by a same lens and coupled to optical spectrum analyzer (OSA). The setup is shown in Fig. S4.

Fiber spectroscopy

To measure the SPDC spectrum, a 3km-long single-mode fiber (SMF) is inserted in the correlation experimental setup. Due to the group velocity dispersion (GVD) of the SMF, the arrival time difference between the two photons depends on their frequencies [2]. To convert the arrival time difference to wavelength, the calibration curves, i.e., the dependence of the arrival time difference on the wavelength of one of the photons, are calculated for each measurement geometry. To this end, a 6 μm thick $LiNbO_3$ layer is used instead of the MS and the histogram of the arrival time differences is measured for photon pairs whose spectrum is restricted with different filters. For one measurement, a bandpass filter with the center wavelength 1200 nm and the width 50 nm is used and for another one, a longpass filter with the cut-off wavelength at 1150 nm. Each filter gives two data points, corresponding to its edges. The fifth point is the one corresponding to the degenerate wavelength and leading to a zero time delay.

Forward-forward and backward-backward cases. In both forward-forward and backward-backward cases, the bandpass or longpass filter is mounted after collimating photons in each arm. With the bandpass filter mounted, the spectral range of the transmitted photon pairs is from 1175 nm (the blue edge of the filter) to 1201 nm (the wavelength of the idler photon matching the signal at the blue edge). Note that both photons in a pair should be in the transmission range of the filter. Similarly, when the 1150 nm longpass filter is mounted, the spectral range of photon pairs is from 1150 nm to 1228 nm.

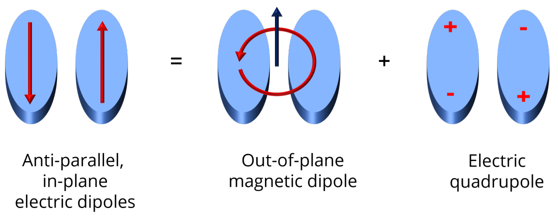
Forward-backward case. In the forward-backward case, the SMF is connected to the arm (transmitted or reflected) whose spectrum has to be measured. To obtain the calibration points, only the bandpass filter is used. When one mounts the bandpass filter at the opposite arm of where the SMF is connected, the spectrum of photons which pass through the fiber is from 1153 nm to 1201 nm. Inversely, when one mounts the bandpass filter at the arm where the SMF is connected, the spectrum is from 1175 nm to 1225 nm.

After getting the data points, the calibration curve is plotted through a fit with a polynomial function (Fig. S7).

The resolution of the SPDC spectrum measurement is 2.76 nm for the forward-forward and backward-backward geometries. For the forward-backward geometry, the resolution is 4.35 nm.

2. SETUPS AND MEASUREMENTS

(a) Bound state in continuum



(b) Quasi-bound state in continuum

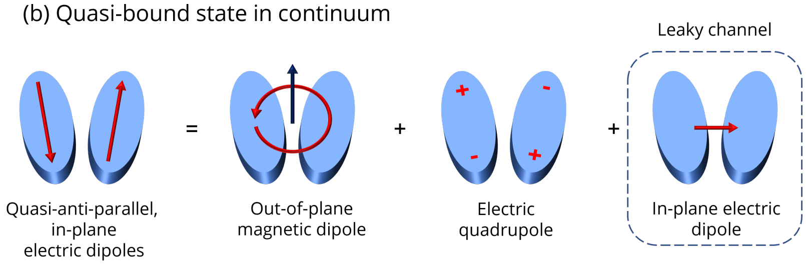


Fig. S1. Schematic depiction of the formation of the (quasi-)bound state in the continuum in the system. (a) A pure bound state in the continuum can be formed when each elliptical particle in the unit-cell supports a single in-plane electric dipole, and they are anti-parallel (i.e., have the same direction but oscillate in anti-phase). In this case, each unit cell (formed by the two ellipses) supports an equivalent out-of-plane magnetic dipole and an electric quadrupole, both having zero net radiation in the out of plane direction. The BIC is formed when all the unit cells are in-phase, a situation in which the only radiation direction allowed is precisely the out-of-plane one. (b) The quasi-BIC is obtained when the symmetry is broken by allowing an angle between the ellipses. In this case, an additional net in-plane electric dipole is supported, opening a leaky channel from which energy can be coupled in and out from the system by normally ingoing and outgoing plane waves with their electric field polarized along the direction of this net dipole.

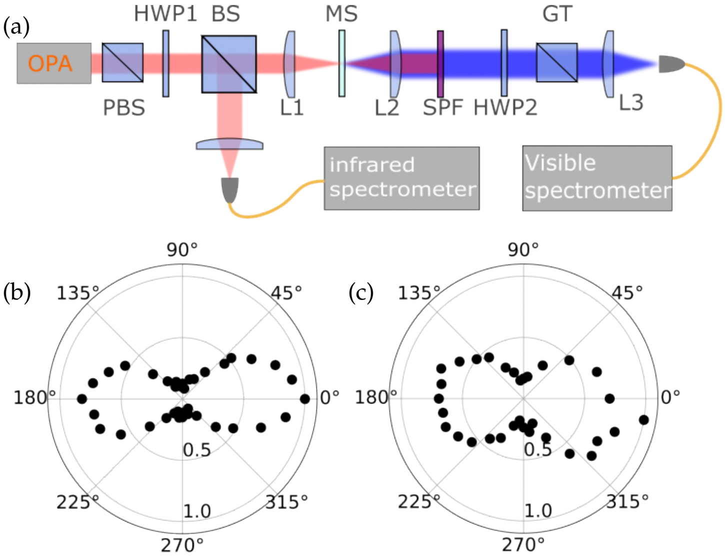


Fig. S2. Polarization dependence of SHG: (a) the experimental setup. The pump beam is horizontally polarized by polarizing beamsplitter PBS. A half-wave plate (HWP1) controls the pump polarization. The pump is split by beamsplitter BS and the reflected beam is sent to an infrared spectrometer. The transmitted beam is focused on the metasurface (MS) by lens L1, with 50 mm focal length. The generated second harmonic radiation is collected by lens L2. A 950 nm short-pass filter (SPF) filters out the pump. The analyzer consists of a half-wave plate (HWP2) and a Glan-Thompson prism (GT). The harmonic signal is sent to a visible spectrometer. (b) The SHG intensity versus the pump polarization. (c) The second harmonic radiation polarization dependence for horizontally (0°) polarized pump.

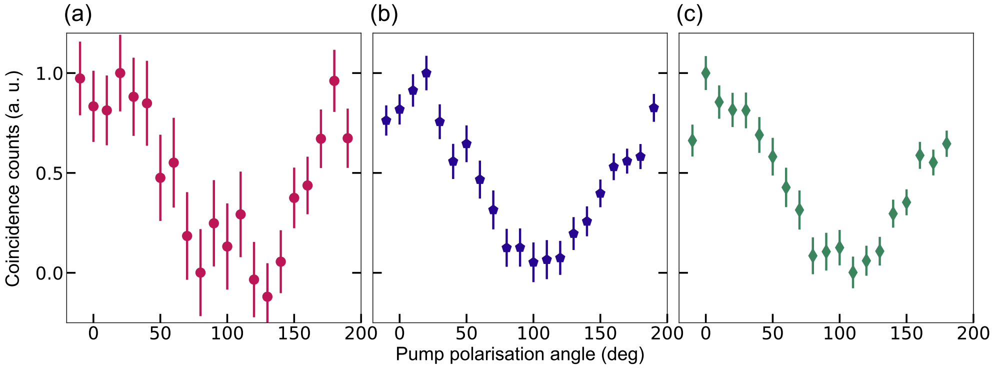


Fig. S3. Normalized SPDC rate measured from the metasurface as a function of the pump polarization angle, with 0° corresponding to the horizontal polarization, for (a) forward-forward, (b) forward-backward, and (c) backward-backward geometries.

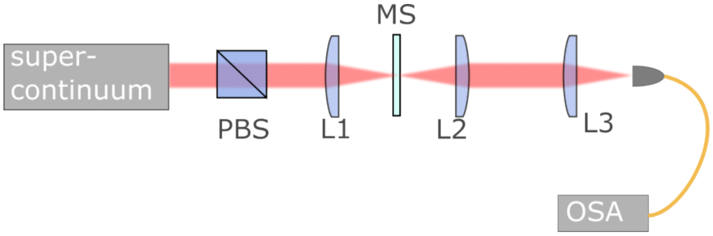


Fig. S4. White-light transmission spectrum measurement setup. The supercontinuum (1kHz repetition rate and 20ps pulse duration) is horizontally polarized by polarizing beamsplitter PBS and focused on the MS by lens L1. The transmitted radiation is collimated by lens L2. Both L1 and L2 are C-coated achromatic lenses with 100 mm focal lengths. The transmitted beam is then coupled into a multimode fiber by lens L3 (focal length 100 mm) and sent to an optical spectrum analyzer (OSA).

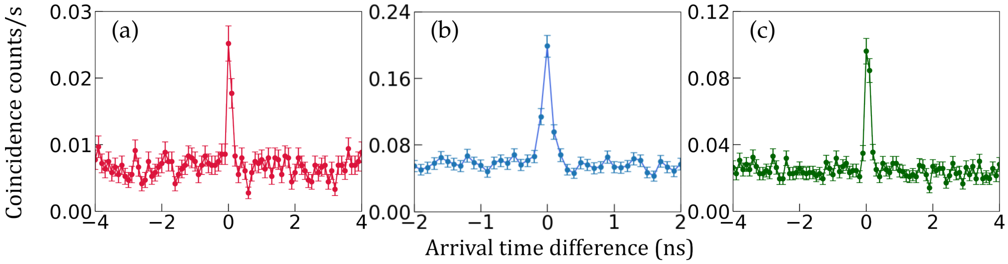


Fig. S5. Coincidence histograms of the MS for (a) forward-forward, (b) forward-backward, and (c) backward-backward measurement geometries. The pump powers are 70 mW for (a) and (c), and 74 mW for (b).

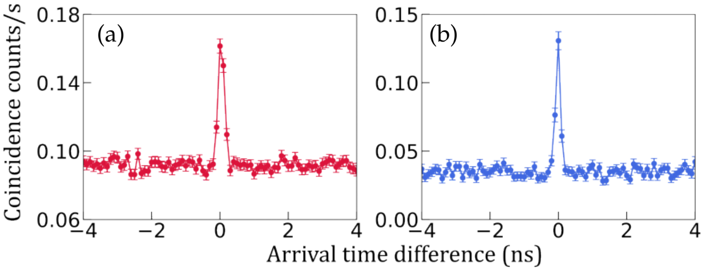


Fig. S6. Coincidence histograms of a 400 nm thick unpatterned GaP film for (a) forward-forward and (b) forward-backward emitted photon pairs. The pump powers are 65 mW for both cases. The time bin width for all measurements is 100 ps. Because of the fiber beamsplitter, half of the coincidence counts in the forward-forward case are lost, which is not accounted for.

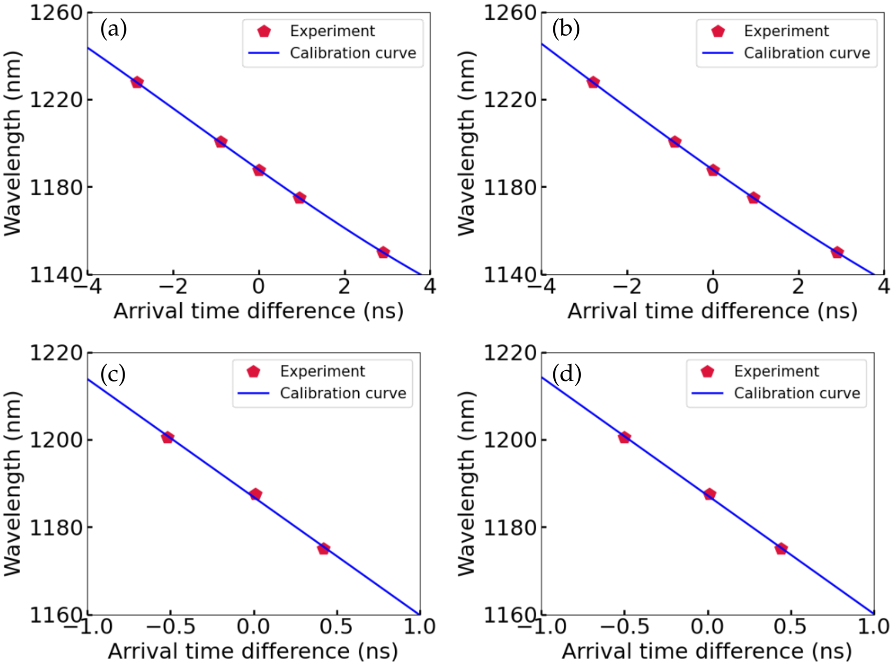


Fig. S7. Calibration curves for the following cases: (a) forward-forward, (b) backward-backward, (c) forward-backward with the fiber in the transmission arm, and (d) forward-backward with the fiber in the reflection arm.

REFERENCES

1. Cameron Okoth. *Spontaneous parametric down conversion on the microscopic scale*. PhD thesis, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 2021.
2. Tomás Santiago-Cruz, Anna Fedotova, Vitaliy Sultanov, Maximilian A Weissflog, Dennis Arslan, Mohammadreza Younesi, Thomas Pertsch, Isabelle Staude, Frank Setzpfandt, and Maria Chekhova. Photon pairs from resonant metasurfaces. *Nano Letters*, 21(10):4423–4429, 2021.