

Supplementary Information for

Counter-propagating Entangled Photon Pairs from a Monolayer

Zhuoyuan Lu^{1,2#}, Jiri Janousek^{1,2#}, Syed M. Assad^{2,3,4}, Shuyao Qiu¹, Mayank Joshi^{2,3,4},
Yecheng Hu⁵, Alex Y Song⁵, Chuanyu Wang¹, Manuka Suriyage¹, Jie Zhao^{2,3}, Ping Koy
Lam^{2,3,4} and Yuerui Lu^{1,2*}

¹School of Engineering, College of Engineering, Computing and Cybernetics, the Australian National University, Canberra, ACT, 2601, Australia

²ARC Centre for Quantum Computation and Communication Technology, the Australian National University, Canberra, ACT, 2601, Australia

³Research School of Physics, College of Science, the Australian National University, Canberra, ACT, 2601, Australia

³ARC Centre for Quantum Computation and Communication Technology, the Australian National University, Canberra, ACT, 2601, Australia

⁴Quantum Innovation Centre (Q.InC), Agency for Science Technology and Research (A*STAR), 2 Fusionopolis Way, Innovis #08-03, Singapore 138634, Republic of Singapore

⁵School of Electrical and Computer Engineering, the University of Sydney, Sydney, New South Wales 2006, Australia

These authors contributed equally to this work.

* To whom correspondence should be addressed: Yuerui Lu (yuerui.lu@anu.edu.au)

Supplementary Note 1: Theoretical modelling of frequency-angular spectrum

Our theoretical predictions expand upon well-established model from previous research^{1,2}. Here, we assume a Gaussian pump mode with frequency ω_p and optical power P_p , with a beam waist radius w_0 . The nonlinear material is characterized by its dispersive refractive index $n(\omega)$ ³, nonlinear susceptibility $\chi^{(2)}$, and length L . The signal and idler wavevectors can be represented as $\mathbf{k}_{s,i} = \mathbf{k}_{s,i\parallel} + \mathbf{k}_{s,i\perp}$, where $\mathbf{k}_{\parallel}$ is the component parallel to the pump direction, and $\mathbf{k}_{\perp}$ is the component perpendicular to the pump propagation. The photon-pair coincidence rate, incorporating experimental parameters such as the signal and idler frequencies $\omega_{s,i}$, emission polar angles $\theta_{s,i}$, and azimuthal angles $\varphi_{s,i}$, is given by¹ :

$$\begin{aligned}
 R_{CC} &= \int_{BW} \int_{\Delta\theta_i} \int_{\Delta\theta_s} \int_0^{2\pi} \int_0^{2\pi} \frac{1}{4 (2\pi)^4} \frac{P_p L^2 w_0^2}{c^7 \epsilon_0 n_p} \chi^{(2)^2} \omega_i^3 (\omega_p \\
 &\quad - \omega_i)^3 n_i^3 n_s^3 \text{sinc}^2 \left[\frac{\Delta k_{\parallel} L}{2} \right] \exp \left[-\frac{(\Delta k_{\perp} w_0)^2}{2} \right] \sin \theta_i \sin \theta_s d\omega_i d\theta_i d\theta_s d\varphi_i d\varphi_s (1)
 \end{aligned}$$

where

$$\Delta k_{\parallel} = \left(\frac{\omega_p n_p}{c} - \frac{(\omega_p - \omega_i) n(\omega_p - \omega_i)}{c} \cos \theta_s - \frac{\omega_i n(\omega_i)}{c} \cos \theta_i \right)$$

$$\Delta k_{\perp}^2 = \left(\frac{(\omega_p - \omega_i) n(\omega_p - \omega_i)}{c} \sin \theta_s \sin \varphi_s + \frac{\omega_i n(\omega_i)}{c} \sin \theta_i \sin \varphi_i \right)^2 + \left(\frac{(\omega_p - \omega_i) n(\omega_p - \omega_i)}{c} \sin \theta_s \cos \varphi_s + \frac{\omega_i n(\omega_i)}{c} \sin \theta_i \cos \varphi_i \right)^2$$

is the longitudinal and transverse wavevector mismatch, respectively. The integration is performed over the idler frequency detection bandwidth (BW) with the signal frequency defined as $\omega_s = \omega_p - \omega_i$. It also spans the detection polar angles $\Delta\theta_{s,i}$, which are set by the numerical aperture of the collection lens and covers photon-pair emission into the full 2π range of azimuthal angles.

Supplementary Note 2: Details of the SPDC experimental setup

A 1550 nm continuous-wave laser (NKT Koheras Adjustik with a 2W Boostik amplifier) was directed into a bow-tie cavity locked on resonance to the 1550 nm laser. The cavity contained a 15mm long periodically poled potassium titanyl phosphate (PPKTP) crystal to generate a 775 nm laser through SHG from the 1550 nm pump. The frequency-doubled output was coupled into a PM optical fiber and served as a pump for the SPDC experiments.

In this work, we employed two optical setups to investigate SPDC in monolayer GaSe. The first setup (Main text Fig. 2b) was designed to measure the co- and counter-propagating coincidence rates of SPDC photon pairs, while the second setup (Main text Fig. 3b) was used for the frequency spectrum and quantum tomography measurements in the counter-propagating regime.

In the first experiment, the 775 nm fiber-coupled pump beam was collimated (using TC25FC-780), transmitted through a dichroic mirror (Thorlabs DMSP950), and focused onto the GaSe

sample with a high numerical aperture ($NA=0.54$) aspheric lens (Thorlabs AL2520M-C). A series of lenses adjusted the beam waist and focus, achieving a $10\text{ }\mu\text{m}$ pump beam waist diameter in the sample. Forward propagating SPDC photons (ie propagating in the direction of the pump beam) were collected with an identical aspheric lens (Thorlabs AL2520M-C), and two 1400 nm long-pass filters (Thorlabs FELH1400) blocked the pump laser. A broadband 50/50 beam splitter (Thorlabs BSW12) was used for correlation measurements, with both reflected and transmitted beams coupled into a pair of multimode fibers (Thorlabs M115L02) via mode-matching optics and 1550 nm fiber couplers (F240FC-1550). Backward-propagating SPDC photons were collected through the same aspheric lens used for focusing the pump beam, reflected off the dichroic mirror, filtered to remove residual pump light, and coupled into another multimode fiber using identical optical components. For photon detection, two SPAD modules (PDM-IR, Micro Photon Devices) were used, and the time-correlated single-photon counting (TCSPC) system (HydraHarp 400, PicoQuant) was connected to both SPADs to process the photon pair correlations.

The SPDC frequency spectrum measurement and quantum state tomography (QST) measurement for the GaSe monolayer was conducted in the counter-propagating regime, as this setup provides twice the probability of detecting photon pairs compared to the co-propagating regime (see main text for details). The experimental setup was similar, with the 50/50 beamsplitter removed, allowing all forward-propagating SPDC photons to be coupled into a single multimode fiber. This configuration maximizes efficiency and accommodates the use of only two SPAD detectors available for the experiment. To measure the frequency spectrum, we placed fiber-coupled tunable bandpass filters (Spectrolight Mono, $1475\text{-}1700\text{ nm}$) before each detector, setting each measurement at a specific wavelength with a bandwidth of 10 nm . To verify polarization entanglement of the generated photon-pairs, we performed QST using identical polarization optics in each arm: a quarter-wave plate (Thorlabs WPQ10M-

1550), a half-wave plate (Thorlabs WPH10M-1550), and a broadband linear polarizer (Edmund Optics).

Supplementary Note 3 | Planar Fresnel Lens Design for Efficient Fiber-Coupled SPDC

Photon Collection

SPDC in monolayer GaSe produces photon pairs emitted across nearly the full 360° in-plane, yielding highly divergent emission. To harness this broad angular span for efficient collection, we designed a planar Fresnel-lens collimator that shapes the SPDC output into a narrow cone, fully compatible with fiber-coupled quantum photonic systems.

Supplementary Fig. 5a illustrates the core concept. The top panel shows a single-side Fresnel lens patterned on a dielectric substrate, collimating SPDC emission (purple arrows) from a monolayer GaSe placed at the rear interface. The collimated emission region is highlighted in green. The bottom panel proposes a fully fiber-integrated geometry, in which two opposing Fresnel lenses and a monolayer GaSe are co-aligned at a shared focal plane. This design supports both unidirectional and bidirectional pumping (shown here with a red arrow for unidirectional pumping) and enables efficient collection of co- and counter-propagating SPDC photon pairs *via* the lens–fiber interfaces, preserving the forward–backward emission symmetry characteristic of monolayer systems.

The Fresnel lenses consist of concentric subwavelength grooves (Supplementary Fig. 5b), ensuring constructive interference and wavefront shaping. Groove depths taper from 270 nm at the center to 200 nm at the periphery (Supplementary Fig. 5c), providing a binary-phase

profile optimized for broadband collimation. This fully planar configuration is scalable and fabrication-compatible with standard dielectric platforms.

We conducted full 3D FDTD simulations using in-plane dipole sources placed at the monolayer GaSe interface to emulate SPDC emission. As shown in Supplementary Fig. 5d–f, the simulated far-field angular emission patterns demonstrate a dramatic narrowing of emission when the Fresnel lens is present (solid curves), compared to the bare substrate (dashed curves), across a wide spectral range from 800 nm to 2000 nm. At 1550 nm, the Fresnel lens achieves over 20× collection enhancement within a $\pm 3^\circ$ detection cone (Supplementary Fig. 5g) compared to the case without lens. This enhancement remains above 15× throughout the entire telecom C-band (1.3–1.6 μm), validating its broadband applicability.

Supplementary Fig. 5h–i show the normalized near-field $|E_x|$ intensity along the optical axis. With the Fresnel lens (Supplementary Fig. 5i), the field exhibits a quasi-flat-top profile, maintaining high intensity over >1 mm propagation length, indicative of low-divergence collimation. Polar plots of far-field intensity at 1550 nm (Supplementary Fig. 5j–k) further highlight the strong directional confinement introduced by the Fresnel design.

The symmetric dual-lens structure proposed in the bottom panel of Supplementary Fig. 5a is compatible with opposing optical fiber interfaces, supporting plug-and-play quantum photonic architectures. It enables simultaneous collection of SPDC photon pairs emitted in both forward and backward directions, preserving the inherent spatial symmetry of monolayer-based SPDC without introducing cavities or resonators. This makes the design particularly attractive for future quantum communication platforms requiring high brightness, broad bandwidth, and fiber compatibility.

Supplementary Note 4: Estimation of detection efficiency of SPDC experiments

In the co- and counter-propagating SPDC setup, the total photon-pair detection efficiency can be estimated as:

$$\eta_{tot} = \eta_{s,i}^2 \eta_{det}^2 \eta_{BS} \eta_{BW} \eta_{Ang},$$

where $\eta_{s,i} = 0.67$ is the efficiency for the signal and idler photons, accounting for all potential optical losses from various components, such as Fresnel losses at the substrate and GaSe sample, collection lenses, long-pass filters, mode-matching optics, fiber couplers, and multi-mode fiber transmission. We neglected any possible interference effects, and we assumed optimal coupling into the multi-mode fibers. The term $\eta_{det} = 0.087$ is the quantum efficiency of the MPDs over the detection bandwidth of 1460–1650 nm, achieved at an excess bias of 2V and the lowest temperature setting. $\eta_{BS} = 0.5$ is the efficiency of the 50/50 broadband beam-splitter used for the co- and counter-propagating photon-pair measurement. The terms η_{BW} and η_{Ang} represent the filtered frequency and angular bandwidth of the SPDC spectrum, respectively. The frequency bandwidth is determined by the MPDs, while the angular bandwidth is defined by the numerical aperture (NA) of the collection lenses. In this setup, the detection angle $\theta_{det} = NA/n_{GaSe} = 0.2$ radians, where $NA = 0.54$ is the numerical aperture of the collection lens and $n_{GaSe} = 2.75$ is the refractive index of the GaSe sample around 1550 nm. For a GaSe monolayer, we estimated $\eta_{BW} = 0.3051$ and $\eta_{Ang} = 0.0037$ relative to the entire frequency-angular spectrum. Similarly, for a 216-layer sample, we estimated $\eta_{BW} = 0.2457$ and $\eta_{Ang} = 0.0112$.

Supplementary Note 5: Maximum-likelihood reconstruction of the density matrix

Quantum state tomography (QST) is a technique used to reconstruct the density matrix of an unknown quantum state based on measurements in multiple bases. While linear inversion can

be used to directly compute a density matrix from measured coincidence counts, this method can yield unphysical results with negative eigenvalues due to statistical noise.

To ensure that the reconstructed density matrix is positive semi-definite, we adopt the maximum likelihood estimation (MLE) method^{4,5}. The MLE approach estimates the most probable physical density matrix that best fits the experimental data by maximizing the likelihood function over a constrained parameter space:

$$\hat{\rho}_{MLE} = \arg \max_{\rho} [\mathcal{L}(\rho)],$$

where the maximization is over all physical states, and the likelihood function is

$$\mathcal{L}(\rho) = Pr(observeddata|\hat{\rho}) = \prod_{j=1}^N \text{Tr}[M_j \rho].$$

Here M_j is the outcome operator corresponding to the j -th click, and N is the total number of clicks.

Iterative method

In practice, the maximum likelihood estimator can be achieved through an iterative expectation-maximisation algorithm

$$\hat{\rho}_{k+1} = \mathcal{N}(R(\hat{\rho}_k) \hat{\rho}_k R(\hat{\rho}_k)),$$

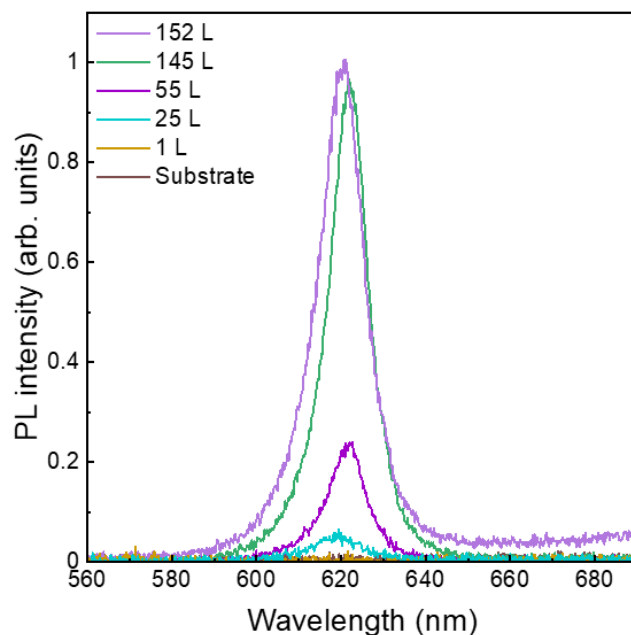
where

$$R(\rho_k) = \sum_{j=1}^N \frac{M_j}{\text{Tr}[M_j \hat{\rho}_k]}$$

and $\hat{\rho}_0$ is an initial guess. The maximum likelihood estimator is obtained after $\hat{\rho}_k$ converges to a stationary point.

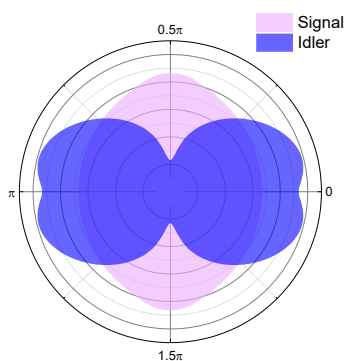
Supplementary Note 6: Polarization dependent Second Harmonic Generation

A 1550 nm continuous-wave laser (NKT Koheras Adjustik with a 2W Boostik amplifier) served as the pump source for second-harmonic generation (SHG). After regulating the power output with a 1550 nm polarizer, the beam was focused onto a 2D thin-film sample using an aspheric lens (Thorlabs AL2520M-C). SHG signals emitted from the sample were collected using an identical aspheric lens and a set of mode-matching optics, then directed to a CCD camera. A 1000 nm short-pass filter was placed before the camera to block the pump wavelength, ensuring only the SHG signal was detected. For polarization dependent SHG measurements, a 1550 nm half-wave plate adjusted the pump polarization before the sample, while a broadband polarizer, aligned with the pump polarization, filtered the SHG signal. By co-rotating the pump polarization and the polarizer, the six-fold rotational symmetry pattern characteristic of the sample's crystal structure was revealed (Supplementary Fig. 9), providing insight into the crystal axes. The armchair and zigzag directions were decided from the six-fold rotational symmetry pattern.

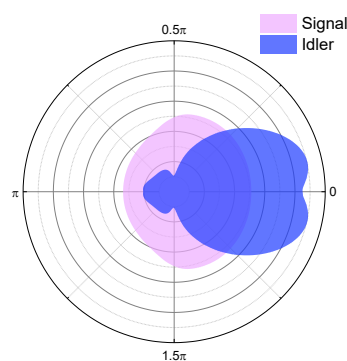


Supplementary Figure 1 | Thickness dependent PL characterization of GaSe. Room temperature PL spectra of GaSe flakes ranging from monolayer (1 L) to 152 L, acquired under a continuous wave 532 nm excitation laser with 60 μ W incident power. PL intensities are normalized to the 152 layer sample (152 L), whose integrated intensity is set to unity.

a

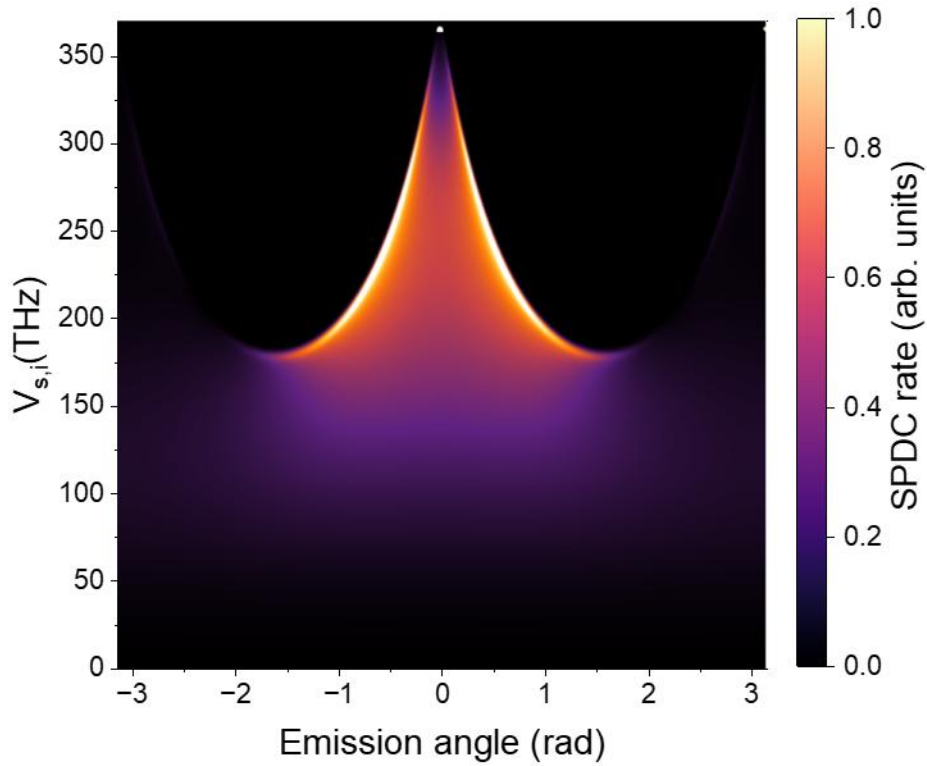


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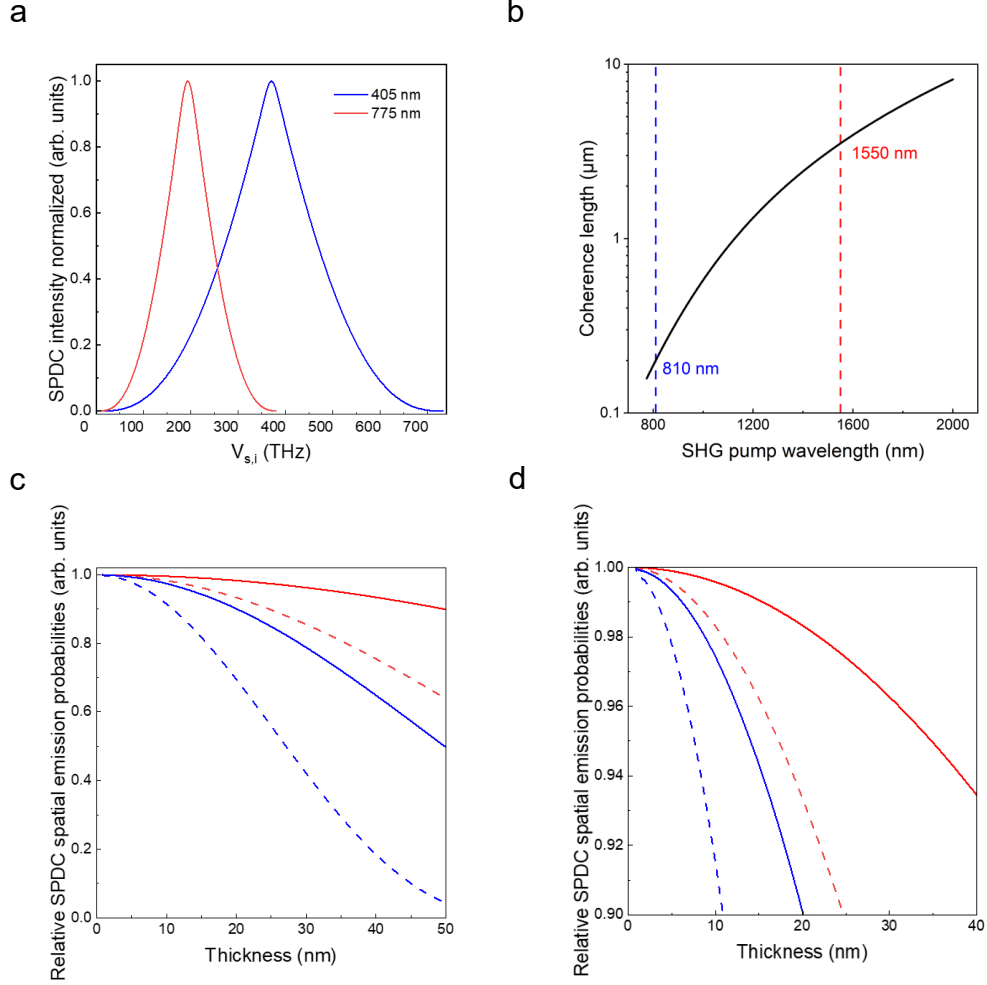


Supplementary Figure 2 | Cross-section of the angular emission spatial profile. a, The cross-section of the simulated angular emission profile for a monolayer GaSe is illustrated in a polar coordinate system, with the pump direction aligned along 0 radians, corresponding to the

forward propagation direction. The sample is positioned at the origin of the polar plot, with its crystal axes oriented perpendicular to the pump direction. **b**, The cross-section of simulated angular profile for a 216-layer GaSe sample.

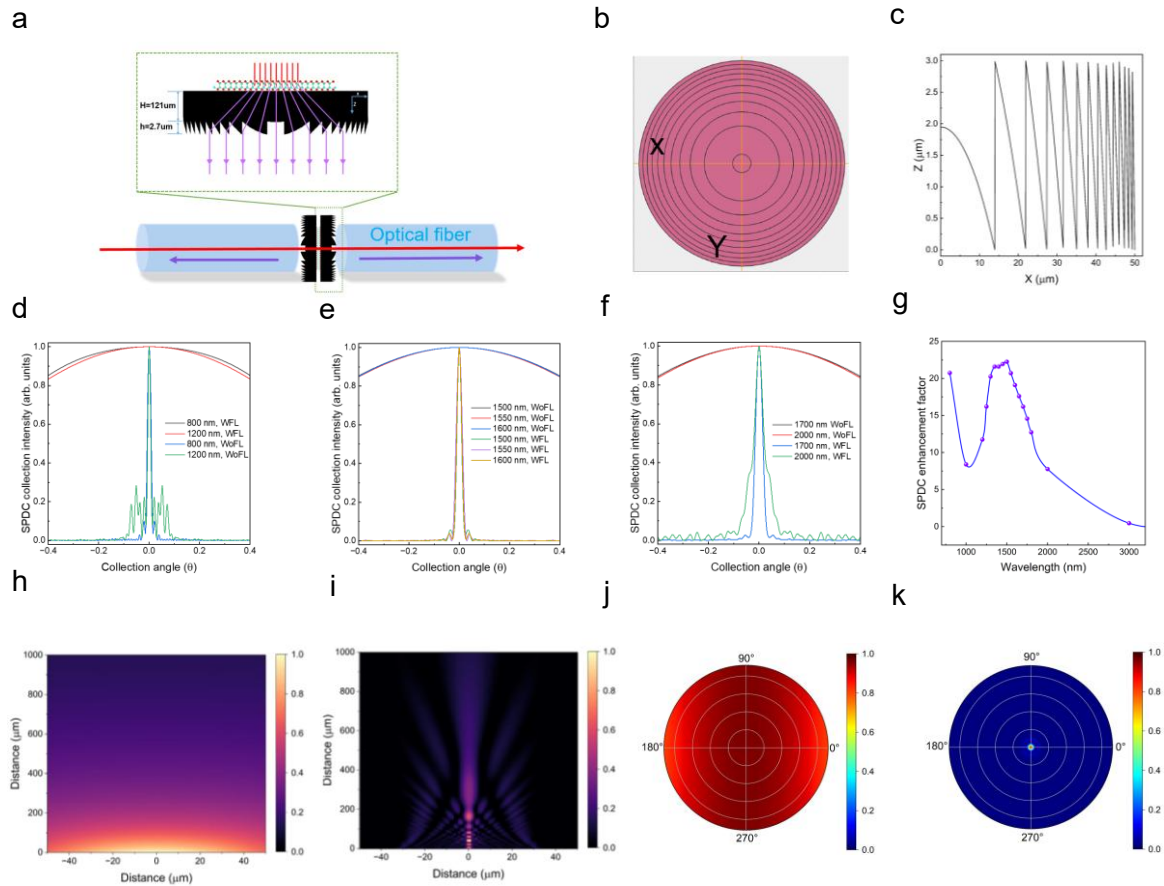


Supplementary Figure 3 | Simulated frequency-angular spectrum of SPDC from a 216-layer GaSe sample. Numerically calculated frequency-angular spectrum of SPDC emission from a 216-layer GaSe crystal. Compared to the monolayer case, the emission profile becomes highly asymmetric, with most of the photon pairs directed into the forward direction. This angular asymmetry originates from the increased thickness of the nonlinear medium, which introduces a tighter phase-matching constraint and suppresses backward emission. The simulation assumes a pump wavelength of 775 nm and accounts for material dispersion based Sellmeier-derived refractive indices³.



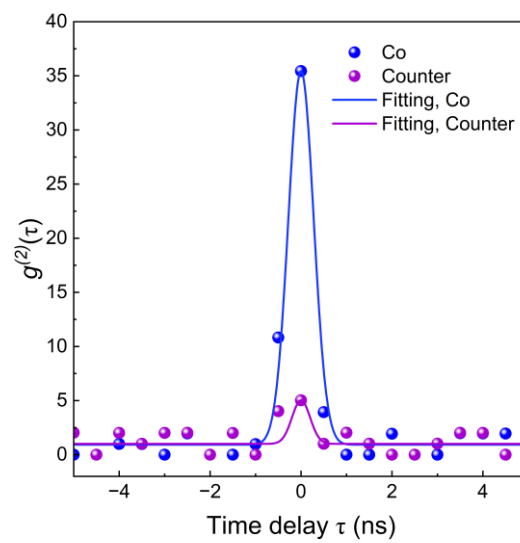
Supplementary Figure 4 | Simulated SPDC spectra, coherence length of GaSe and relative SPDC spatial emission probabilities. **a**, Normalized SPDC frequency spectra from a GaSe monolayer under our experimental conditions, shown for 775 nm (red) and 405 nm (blue) pump wavelengths. For the 775 nm pump, the SPDC bandwidth $\Delta\nu$ is approximately 100 THz, corresponding to a temporal correlation time of $\tau_c = \frac{1}{\pi\Delta\nu} \approx 3.2$ fs, assuming a Lorentzian emission spectrum. **b**, Calculated wavelength-dependent coherence length of GaSe based on Sellmeier-derived refractive indices³. **c**, Relative SPDC spatial emission probabilities with increasing GaSe film thickness. The solid curves show the normalized probability of biphoton emission for scenario (II), in which the signal photon is emitted forward and the idler backward, normalized to scenario (I), where both photons are emitted forward. Due to symmetry, scenario (III)—the reverse of scenario (II)—has the same probability and is represented by the same curve. The dashed curves show the normalized probability for scenario (IV), where both photons are emitted in the backward direction, also referenced to scenario (I). Red: 775 nm SPDC excitation, Blue: 405 nm SPDC excitation. **d**, Enlarged view of panel (c), focusing on

the regime where relative emission probability ratios approach unity. This zoomed-in region highlights the sensitivity of the emission probability ratio to small variations in GaSe thickness across different SPDC scenarios.

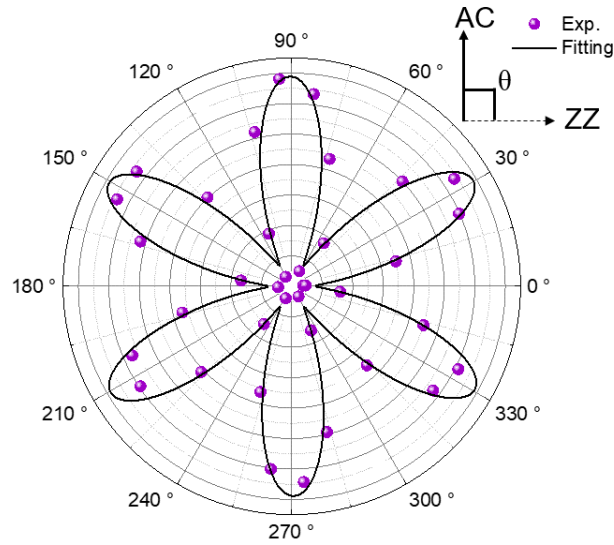


Supplementary Figure 5 | Fresnel collimating lens design for SPDC and its collection performance. **a**, Optical schematic illustrating SPDC collimation and integration schemes using Fresnel lenses. Top: A single-side Fresnel lens patterned on a dielectric substrate collimates SPDC emission (purple arrows) from a monolayer GaSe placed on the rear face. The green box highlights the collimated emission region. Bottom: A conceptual design of a fiber-integrated platform with two opposing Fresnel lenses and a monolayer GaSe located at their shared focal plane. The 775 nm pump beam (red arrow) generates co- and counter-propagating SPDC photon pairs, which are efficiently collected via the integrated lens-fiber interfaces. **b**, Plan view of the concentric-ring groove pattern. **c**, Cross-sectional profile of the grooves, with depths tapering from 270 nm at the innermost ring to 200 nm at the outermost. **d–f**, Far-field E-field intensity distributions at $\phi = 0$ plane as a function of polar angle θ for

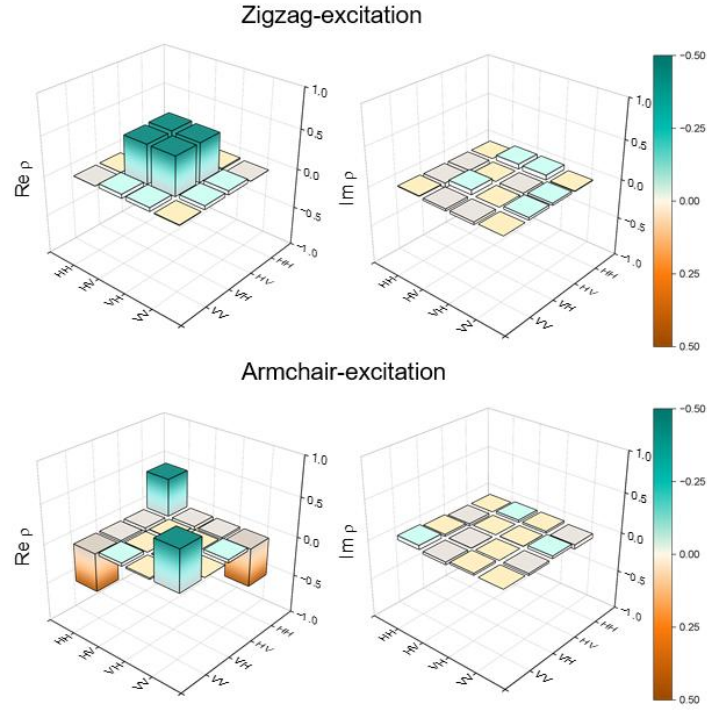
wavelengths of 800 nm and 1200 nm (d), 1500 nm, 1550 nm and 1600 nm (e), and 1700 nm and 2000 nm (f), comparing configurations with and without the lens (solid and dashed lines, respectively). **g**, Broadband enhancement of SPDC within $\pm 3^\circ$ about the surface normal, plotted as a function of wavelength from 700 nm to 3000 nm. **h,i**, Normalized near-field $|E_x|$ intensity profiles at 1550 nm along the optical axis from 0 to 1000 μm without (h) and with (i) the Fresnel lens. **j,k** Polar plots of normalized far-field angular intensity at 1550 nm without (j) and with (k) the Fresnel lens; all curves are peak-normalized to unity.



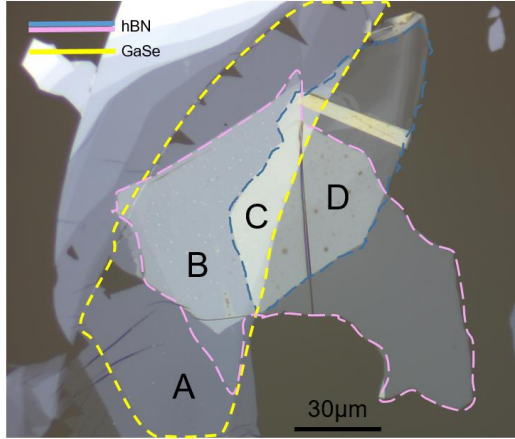
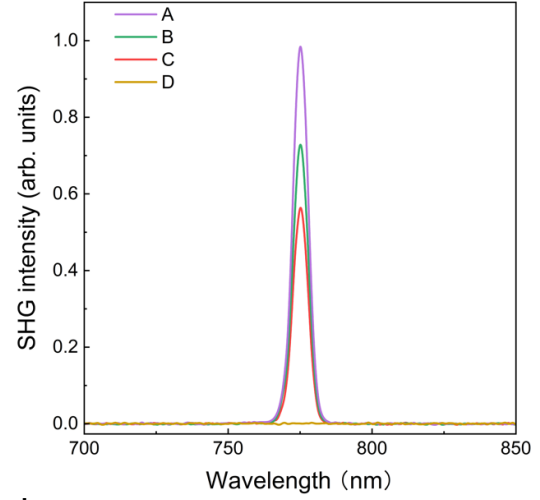
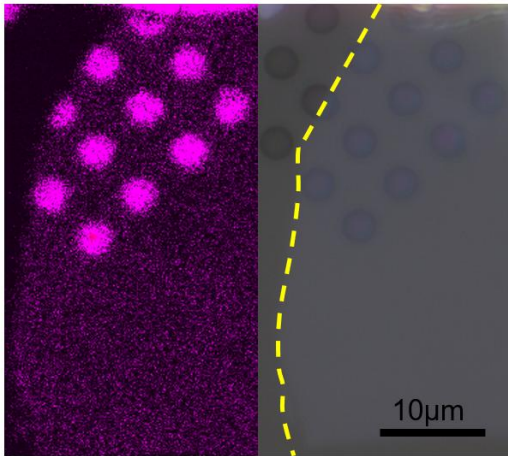
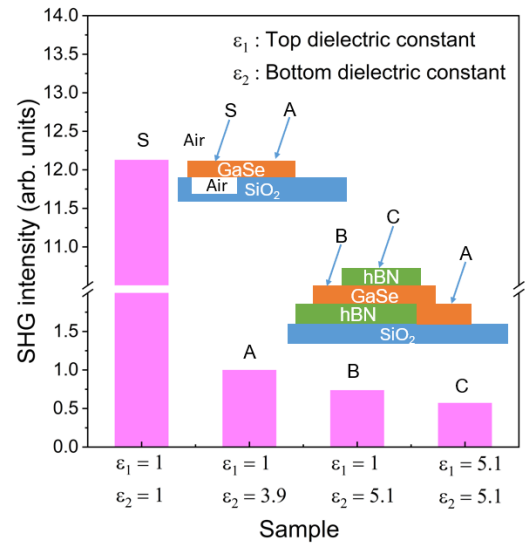
Supplementary Figure 6 | Normalized second order correlation function $g^{(2)}(t)$ for the 216-layer GaSe in co-propagating and counter-propagating scenarios, with average excitation power $P = 30$ mW and integration time of 240 s and 0.5 ns bin width.



Supplementary Figure 7 | Polarization dependent SHG from GaSe sample. The six-fold symmetric pattern is preserved from monolayer to bulk crystal. Maximum SHG was observed along the armchair direction (AC), and minimum SHG along the zigzag direction (ZZ). To determine the crystal axis of a monolayer, we first prepared a sample where the monolayer was attached to a bulk region sharing the same crystalline orientation. Polarization-dependent SHG measurements were conducted on the bulk region to serve as a reference for identifying the crystal axis of the attached monolayer. This reference was then used to guide the QST measurements on the monolayer.



Supplementary Figure 8 | Quantum state tomography for a 90-layer sample with counter-propagating collection, with the pump aligned along the armchair (AC) and zigzag (ZZ) directions. Results show 98% fidelity to the expected Bell states: with pump polarization along the AC orientation, the state $|\Phi^-\rangle = 1/2(|HH\rangle - |VV\rangle)$ is produced, and along the ZZ orientation, the state $|\Psi^+\rangle = 1/2(|HV\rangle + |VH\rangle)$ is generated.

a**b****c****d**

Supplementary Figure 9 | Optical characterization and SHG measurements of hBN-encapsulated and suspended GaSe. **a**, Optical microscope image of a few-layer GaSe sample partially encapsulated by hBN layers. The yellow dashed line outlines the GaSe region, while the pink and green dashed lines mark the top and bottom hBN layers, respectively. Four regions are labeled: A (GaSe/SiO₂), B (GaSe/hBN/SiO₂), C (hBN/GaSe/hBN/SiO₂), and D (hBN/hBN/SiO₂ as a reference region). **b**, Comparison of SHG intensities measured from regions A, B, C, and D under 1550 nm pulsed laser excitation, highlighting the impact of

different encapsulation configurations on the nonlinear optical response of GaSe. **c**, Left: Optical microscope image showing a few-layer GaSe flake suspended over a free-standing circular hole with a diameter of 3 μm and a depth of 3 μm . Right: Corresponding SHG mapping acquired under 900 nm pump excitation, visualizing the SHG emission from the suspended and substrate-supported regions. **d**, Quantitative comparison of SHG intensities across different dielectric environments. The bar chart summarizes the normalized SHG intensities for GaSe under four configurations: S: Suspended GaSe ($\epsilon_1 = \epsilon_2 \approx 1$); A: GaSe/SiO₂ ($\epsilon_1 \approx 1$, $\epsilon_2 \approx 3.9$); B: GaSe/hBN ($\epsilon_1 \approx 1$, $\epsilon_2 \approx 5.1$); C: hBN/GaSe/hBN ($\epsilon_1 \approx 5.1$, $\epsilon_2 \approx 5.1$), where ϵ_1 and ϵ_2 are the dielectric constants of the layers on top of and underneath GaSe, respectively. The SHG intensity of the GaSe/SiO₂ configuration is normalized to 1.

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