

Supplementary Materials for

Quantum computational advantage using photons

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1 Two-Mode Squeezed State (TMSS) Sources

This section provides a detailed description of the TMSS sources.

1.1 The pumping laser system

Layout of the laser systems used in this experiment is shown in Fig. S2A. The system employs a mode-locked Ti:Sapphire oscillator (Coherent Mira 900) pumped by a semiconductor laser (Coherent Verdi G) at 532 nm, which generates pulsed seed laser with a repetition rate of 76 MHz and a central wavelength of 776 nm. Before entering the ultra-fast amplifier (Coherent RegA 9000), the pulsed seed laser first goes through a Stretcher/Compressor module to be expanded in time domain, so that the peak intensity is low to avoid damage on the amplifying medium and minimize nonlinear distortion of the pulse. For flexible wavelength control and precise dispersion compensation, the seed laser is then directed into an external spectrum shaping subsystem, which consists of transmission grating and lens for spectrum modification, as shown in Fig. S2B. In this way, the central wavelength and linewidth of the seed laser can be adjusted by the position and width of the slanted slit. Furthermore, we exploit a 140-element deformable mirror to adaptively control the phase between different wavelength components, which compensates high-order dispersion effectively. After spatial filtering, each wavelength is then combined through another transmission grating. The modified seed laser is then fed into the regenerative amplifier (RegA 9000), which is pumped by another Verdi G at 532 nm. Based on the chirped pulse amplification technique, RegA 9000 boosts the single pulse energy to micro-joule level and reduces the repetition rate to 250 kHz. In the final step, the pulses exit the laser system after being compressed into ultra-short pulses with widths of 200 fs.

To monitor the quality of the ultrafast laser pulses in real time, we adopt a method called frequency-resolved optical gating (FROG). Compared to the auto-correlation method, FROG enables us to directly read out intensity and phase information of the spectrum, providing a reference for dispersion compensation. In our experiment, we evaluate output of the laser system with Swamp Optics GRENOUILLE 8-50-USB. The intensity and phase

of the spectrum are shown in Fig. S3, indicating that pump beam of the TMSS sources is centered at 776 nm, and the phase deviation is reduced to less than 0.002 rad in the range from 770 nm to 780 nm by adjusting the dispersion compensation module.

1.2 Design of the periodically poled KTiOPO₄ (PPKTP) crystal

We adopt a 4-mm PPKTP crystal with a poling function as shown in Fig. S4 (the blue line). The design brings about two advantages. Firstly, near the center of the crystal where the major PDC photon's yield is contributed, the poling period perfectly satisfies phase matching condition, corresponding to an maximum PDC yield. Secondly, as moving to both ends of the crystal, the periodicity is modulated in a way that the PDC photon's yield rate follows a Gaussian function, dropping to 0 at both ends. The joint spectrum, which is the Fourier transform of the Gaussian photon yield, is thus also Gaussian without sidelobes (*36*). The red circles in Fig. S4 show the simulated amplitude of down-conversion beam as an function of crystal length, which is consistent with an ideal Gauss error function (the green line).

1.3 Experimental setup of the TMSS sources

In this work, 25 TMSS sources are required to arrive at the interferometer simultaneously. Here, we adopt a compact optical layout as shown in Fig. S5. The pumping laser firstly goes through a pair of cylinder lenses to reform the beam's profile into a Gaussian shape. The pumping laser is then separated into 13 paths with equal intensity by a series of polarization beam splitters and half-wave plates. Pumping laser in each path is initialized to horizontal polarization before sent to the crystal for phase-matching. For 12 out of the 13 paths, there are two crystals pumped in a cascading configuration. The laser is tightly focused on the crystals with a beam waist of 60 μ m. The generated 1552 nm down-converted photons are then separated from the 776 nm pumping laser with a dichroic mirror (DM), which reflects 1552-nm photons and transmits 99% of the pump beam. The down-converted photons and the remaining 1% of the pump beam are then collected by single-mode fibers (CORNING SMF-28 Ultra). For optimal match from free-space to the

single-mode fibers, 100-mm-f plano-convex lenses together with collimators (NA=0.25) are utilized in each path. Before each collimator, 2-mm KTP crystal is placed with optical axis orthogonal to the PPKTP crystal to compensate temporal mismatch ($\sim 190 \ \mu m$) between the horizontally polarized (H) and vertically polarized (V) photons due to bire-fringence of the PPKTP crystal. All the collimators are attached to one linear translation stage along the light propagation direction, which can be adjusted to ensure wave packages of 25 TMSSs overlapped perfectly in the interferometer.

The refractive index of the PPKTP crystal changes with temperature, which accordingly changes the phase-matching condition. As shown in Fig. S6, we adjust the temperature of all the crystals into the degenerate point. In our setup, all the PPKTP crystals are independently PID-controlled with thermoelectric cooler (TEC).

1.4 Measurement of the joint spectrum and spectral purity with threshold detectors

We measure the joint spectrum by first converting the frequency information into temporal information with a long dispersion fiber, and then using a time-to-digital converter (TDC) to time tag the coincidence between the signal and idler photon. The obtained time tags of the coincidence, relative to a ~ 250 kHz synchronization signal from the pumping laser, thus yields information of the frequency of the signal and idler photon respectively after a dispersion calculation. In Fig. S7 (Fig. 2C) we show the joint spectrum of the photon pair after (before) 12-nm filtering, which corresponds to a spectral purity of 0.99 (0.98). We also use a spectrometer to measure the spectrum for each polarization as shown in Fig. S8.

The spectral purity of the signal and idler photon is an important metric for multiphoton interference. We use unheralded second-order correlation measurement (24) as a quantitative measure of the purity of the TMSS source. Following Ref. (24), the purity of the photon detected with ideal detectors is $\mathbb{P} = g^{(2)}(0) - 1$. The experiment used threshold single photon detectors which is not photon-number-resolving, so we need to correct the measured unheralded second order correlation. The spectral purity can be defined as (24)

$$\mathbb{P} = \sum_{i} \lambda_i^4 \tag{1}$$

where λ_i is the mode distribution of the *i*-th mode, with $\sum_i \lambda_i^2 = 1$. The photon-numberresolving $g^{(2)}(0)$ measurement yields

$$g^{(2)}(0) = 1 + \frac{\sum_{i} \sinh^{4}(r_{i})}{(\sum_{i} \sinh^{2}(r_{i}))^{2}}$$
(2)

Because tracing out one port of the TMSS yields a thermal state (34), one can rewrite (2) in the form of

$$g^{(2)}(0) = 1 + \frac{\sum_{i} \bar{n}_{i}^{2}}{(\sum_{i} \bar{n}_{i})^{2}}$$
(3)

where \bar{n}_i is the average photon number of the *i*-th mode with $\bar{n}_i = \sinh^2(r_i)$.

In this experiment, it is performed as a Hanbury Brown and Twiss (HBT) type measurement of one port of the TMSS source. As our source is spectral-correlation-free, we can model the state of one port of the TMSS as a mixing of the major mode with average photon number \bar{n} , and a minor mode with average photon number \bar{m} , satisfying $\bar{m} \ll \bar{n}$.

$$\rho = \rho_{\rm th}(\bar{n}) \otimes \rho_{\rm th}(\bar{m}) \tag{4}$$

The probability of causing a coincidence count at t = 0, by having the two modes as inputs, can be calculated as

$$P(0,\bar{n},\bar{m}) = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \left[\left(1 - \frac{1}{2^{m+n-1}}\right) \times \frac{1}{1+\bar{n}} \left(\frac{1}{1+\bar{n}}\right)^n \times \frac{1}{1+\bar{m}} \left(\frac{\bar{m}}{1+\bar{m}}\right)^m \right] - \left(1 - \frac{1}{2^{-1}}\right) \frac{1}{1+\bar{n}} \frac{1}{1+\bar{m}} = \frac{2(\bar{n}^2 + \bar{m}^2) + 2\bar{n}\bar{m} + 3(\bar{n}^2\bar{m} + \bar{m}^2\bar{n}) + \bar{n}^2\bar{m}^2}{(1+\bar{n})(2+\bar{n})(1+\bar{m})(2+\bar{m})}$$
(5)

The probability of causing a coincidence count at $t = \tau$, with τ the laser pulse period and

 $\tau \gg \tau_c$ the coherent time of the photon, can be calculated as

$$P(\tau, \bar{n}, \bar{m}) = \left[\sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \left(1 - \frac{1}{2^{m+n}}\right) \times \frac{1}{1+\bar{n}} \left(\frac{1}{1+\bar{n}}\right)^n \times \frac{1}{1+\bar{m}} \left(\frac{\bar{m}}{1+\bar{m}}\right)^m\right]^2 = \frac{(\bar{n}\bar{m} + 2(\bar{n}+\bar{m}))^2}{(2+\bar{n})^2(2+\bar{m})^2}$$
(6)

The experimentally measured $g^{(2)}(0)$ is

$$g^{(2)}(0) = \frac{P(0,\bar{n},\bar{m})}{P(\tau,\bar{n},\bar{m})}$$
(7)

To calculate spectral purity, we also measure the total average photon number in experiment by using the relationship of a threshold detector triggered by a thermal state with average photon number \bar{N}

$$\bar{N} = \frac{p}{1-p} \tag{8}$$

where p is the probability of the detector fires. In our case we have

$$\bar{N} \approx \bar{n} + \bar{m}.\tag{9}$$

With (7), (8) and (9), we solve the average photon number of the major and minor mode from the measured $g^{(2)}(0)$ and p

$$\begin{cases} g^{(2)}(0) = \frac{P(0, \bar{n}, \bar{m})}{P(\tau, \bar{n}, \bar{m})} \\ \frac{p}{1-p} = \bar{n} + \bar{m} \end{cases}$$
(10)

and finally, the purity

$$\mathbb{P} = \frac{\bar{n}^2 + \bar{m}^2}{(\bar{n} + \bar{m})^2}$$
(11)

As an illustration we show in Fig. S9A a plot of the dependence of the measured $g^{(2)}(0)$ on the firing probability of the threshold detectors, with fixed purity $\mathbb{P} = 0.95$. We also show in Fig. S9B a plot of a typical measured $g^{(2)}(0)$ result.

2 100-Mode Large-Scale Linear Optical Network

This section provides detailed description of the 100-mode large-scale interferometer that acts an unitary transformation on the input 25 TMSS sources.

2.1 Fabrication of the interferometer

The required 100-mode linear optical network should simultaneously combines phase stability, full connectivity, matrix randomness, perfect wavepacket overlap and high transmission efficiency. The setup is shown in Fig. 2C in the main text, the optical network consists of a five-layer triangular interferometer and a five-layer rectangular one. Every rectangular (triangular) layer contains 10 (5) input modes, and the equivalent photonic circuit is shown in Fig. S10. We first discuss the fabrication of the interferometer. The working principle will be presented in the next subsection.

The rectangular interferometer is made of 10 trapezoid-shaped fused quartz plates with a size of $50.91 \times 30 \times 4.24 \text{ mm}^3$ (see Fig. S11A). Due to the short coherence length of the TMSS source (~88 µm) utilized in our experiment, a much more stringent requirement on the fabrication precision is required, compared to the previous design (*18, 38*). After each trapezoid was cut and finely polished, we measured the thickness of every plate using a white light interferometry. The measured standard deviation of plate thickness is 98 nm, which is three orders of magnitude smaller than the coherence length of TMSS sources. Very similar process is done to the plates used for triangular interferometer, whose standard deviation is 198 nm, also much smaller than the coherence length of TMSS sources.

Next, the surfaces L2-L10 are optically coated with polarization-dependent beamsplitting thin films with a designed splitting ration of $T_s:R_s=35:65$ for the s wave and $T_p:R_p=75:25$ for the p wave. Because the refractive index of quartz is ~1.45, the light incident at 45 degree will be totally reflected by L1 and L11. Finally, the 10 trapezoids are bond together one-by-one via intermolecular Van der Waals forces, and 20 outer surfaces for both inputs and outputs are antireflectively coated to improve the transmission efficiency. Likewise, the triangular interferometer is fabricated with such a process, of which L1-L4 are polarization-dependent beam-splitting coated, and two outer surfaces for input and output are antireflectively coated to improve the transmission efficiency (see Fig. S11B). The size of the rectangular (triangular) one is $50.91 \times 42.42 \times 30 \text{ mm}^3$ ($30 \times 30 \times 30 \text{ mm}^3$), the whole optical network contains 300 beam splitters and 75 mirrors.

2.2 Working principle of the interferometer

The equivalent photonic circuit of our 3D interferometer is presented in Fig. S10. The TMSS photons first propagate through the triangular interferometer and then the rectangular one. The injected photons in the same triangular layer can talk with each other. Next, five rectangular layers are implemented to let photons in different triangular layer talk with each other (see Fig. S10). Thus a fully-connected 50×50 -mode matrix is generated.

We further add the polarization degree of freedom to realize a 100×100 -mode unitary matrix. There is no interference between H and V modes inside the 50-mode linear optical network independently. The corresponding matrix can be written as

$$U_{2m\times 2m}^{\text{spa}} = \begin{pmatrix} H_{11} & H_{12} & \cdots & H_{1m} & 0 & 0 & \cdots & 0 \\ H_{21} & H_{22} & \cdots & H_{2m} & 0 & 0 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ H_{m1} & H_{m2} & \cdots & H_{mm} & 0 & 0 & \cdots & 0 \\ \hline 0 & 0 & \cdots & 0 & V_{11} & V_{12} & \cdots & V_{1m} \\ 0 & 0 & \cdots & 0 & V_{21} & V_{22} & \cdots & V_{2m} \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 0 & V_{m1} & V_{m2} & \cdots & V_{mm} \end{pmatrix}$$
(12)

where every element $A_{ij} = r_{ij}e^{-i\phi_{ij}}$. To make a fully-connected unitary matrix, we adopt a half-wave plate and a polarized beam splitter (PBS) to every output port of the interferometer as a tunable beam splitter. In this case, H- and V- polarized photons can interference with each other on PBS, as shown in the Fig. S10. The transformation matrix of these 50

PBSs can be written as

$$U_{2m\times 2m}^{\text{pol}} = \begin{pmatrix} p_{1H,1H} & 0 & \cdots & 0 & p_{1V,1H} & 0 & \cdots & 0 \\ 0 & p_{2H,2H} & \cdots & 0 & 0 & p_{2V,2H} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & p_{mH,mV} & 0 & 0 & \cdots & p_{mV,mH} \\ \hline p_{1H,1V} & 0 & \cdots & 0 & p_{1V,1V} & 0 & \cdots & 0 \\ 0 & p_{2H,2V} & \cdots & 0 & 0 & p_{2V,2V} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & p_{mH,mV} & 0 & 0 & \cdots & p_{mV,mV} \end{pmatrix}$$
(13)

at the same basis with the $U_{2m\times 2m}^{\text{spa}}$. Therefore, the matrix $U_{2m\times 2m}^{\text{pol}}U_{2m\times 2m}^{\text{spa}}$ is the final unitary transformation of the linear optical network. In this experiment, *m* value is 50, so we finally have a 100×100 fully-connected unitary matrix.

2.3 Characterization of spatial and temporal overlap

The main function of the interferometer is to realize the perfect overlap of photon wave packets both spatially and temporally. To this end, we have carefully polished the quartz plates to ensure a high-level fabrication precision and a high-level parallelism between two surfaces. Before putting into use, we test the final off-the-shelf interferometer to check if it can work properly.

To calibrate the spatial overlap between any two modes, we use a Mach-Zehnder-type interferometry by splitting a continue-wave laser with a linewidth of \sim MHz into two beams and then fed into two modes of the interferometer. By monitoring the visibility of the fringe pattern at different output ports, the determined average spatial overlap is more than 99.5%. This visibility is observed simultaneously with an average single-mode fiber collection efficiency of more than 92% for all 100 modes. These two results indicate that the spatial overlap of interferometer itself is perfect.

Next, we calibrate the temporal overlap inside the interferometer. The temporal overlap is essential in this experiment, since the coherence length of the TMSS source is only $\sim 88\mu$ m. If there are some serious temporal mismatch in the interferometer, the quantum interference would decrease and the GBS system would decay to a classical one. To determine the temporal overlap, two inferometers are cascaded to form multiple Mach-Zehnder-type interferometers. The path difference is also measured by while light interferometry. After passing through the first interferometer, only two beams are allowed to inject the second interferometer while the other ports are blocked. By monitoring the interference fringe pattern, we can know all path differences between any two of the ports. The measured results of rectangular interferometer are listed in Fig. S12, which indicates that the path differences between any two ports are well below 10 μ m, and its standard deviation is $\sim 2.6 \ \mu$ m. If we map path differences to the calculated Hong-Ou-Mandel visibility curve of the TMSS source (see Fig. S13), the temporal mismatch caused by the interferometer itself will make the visibility drop by 1% at most. The very similar results are obtained from the triangular interferometer at a same precision lever. Therefore, we conclude that the interferometer used in this experiment will make traveling photons have a near-perfect temporal and spatial overlap.

2.4 Experimental setup of the linear optical network system

Since the GBS is sensitive not only to the phase inside the interferometer but also to the phase before the interferometer, we place it on a optical stage as stable as possible. To minimize any drift caused by temperature wandering, all optical elements including four interferometers (a triangular one and a rectangular one, and the other two is for active phase lock), one PBS, one mirror and one dichromatic mirror (light at 1552 nm pass while light at 776 nm reflected), are all adhered to a ultra-low-expansion quartz plate with a size of $200 \times 200 \times 40 \text{ mm}^3$ (see Fig. 2C in the main text and Fig. S14).

Another challenge is the alignment of the light beam and efficiently collect photons into single-mode fibers. There are 25 independent input ports and 100 output ports, resulting 2500 beam alignment. To mount the entire device with a large number of optical elements on a limited optical table, We have to miniaturize each device as compact as possible. A schematic diagram of the device is in Fig. S14A to show the arrangement of all optical elements. A photograph of our interferometer system is shown in Fig. S14B. There

are 25 input modes (shown in red box) and 100 surrounding output modes (shown in blue box). Because the distance between two adjacent optical beams is 6 mm, mini-mirrors with size of $6 \times 6 \times 6 \text{ mm}^3$ are carefully bonded on many different stages to separate all beams to 100 surrounding collimators. Such an architecture can ensure the distances from all 100 output collimators to 25 input collimators are almost same, a necessary condition for high collection efficiency.

2.5 Matrix elements measurement

All amplitude elements are determined by counting thermal-state photons at the end of the single-mode fiber. It's experimentally convenient, since tracing one mode of the TMSS will lead to a leaving thermal state. Experimentally, we can put a polarizer in front of the interferometer to let *H*-polarized mode of TMSS pass through the linear optical network while filtering the *V*-polarized photons, and then detect them using 100 superconducting single-photon detectors. We can easily get the click counts $(c_{i1}, c_{i2}, \dots, c_{i100})$ for the *i*-th input port after a certain accumulating time. However, a thermal state contains uncertain number of photons while we use single-photon detectors to do threshold detection, we need to correct the mean photon number of every port. For a thermal state, the probability of zero-photon case is

$$P(0) = \frac{1}{\bar{n}+1},$$

where \bar{n} is the mean photon number. Therefore, the probability for threshold detection is

$$P_{\text{thres}} = 1 - P(0) = \frac{\bar{n}}{\bar{n}+1},$$

thereby the mean photon number should be

$$\bar{n} = \frac{P_{\text{thres}}}{1 - P_{\text{thres}}}.$$

The final determined amplitude for the *i*-th raw is proportional to

$$\left(\frac{c_{i1}/C}{1-c_{i1}/C}, \frac{c_{i2}/C}{1-c_{i2}/C}, \cdots, \frac{c_{i100}/C}{1-c_{i100}/C}\right),$$

where C is the number of pulses during the measurement. All amplitude elements are shown in Fig. 2E in the main text.

All phase elements are determined by a narrow-band continue-wave laser (*37*). Before sending it into the optical network, we spilt it into two beams. One of them is a reference beam, while the other beam pass through two acoustic optical modulators (AOM) to have a 5 Hz frequency difference with respect to the reference beam. Next, two beams are injected into two input ports of interferometer. At the output port, a clear beat pattern will be observed. By comparing the phase with respect to the reference port, all phase elements can be determined, which are shown in Fig. 2F in the main text. The reconstructed matrix elements of the 100-mode interferometer are demonstrated in Table. S2 and Table. S3.

2.6 Unitary test and randomness of reconstructed matrix

To demonstrate the correctness of the reconstructed matrix elements, a unitary test UU^{\dagger} is applied to it. If the measured elements are consistent (both the amplitudes and phases) with each other, the value UU^{\dagger} must be very close to identity. Fig. S15A (S15B) shows the real (imaginary) part of the unitary test, the average value of all 4900 non-diagonal elements is as small as 0.012 (0.012), thus confirming the high degree of unitarity of the reconstructed matrix.

A necessary condition for the hardness of the GBS is that the unitary matrix should be Haar-random. To this aim, we generate 1000 random unitary matrices according to Haar measure, and then compare the statistical results of both the amplitude and phase elements. Theoretically, the distribution of the amplitude element of Haar unitaries obeys a distribution of $2(m - 1)(1 - r^2)^{m-2}r$ (not normalized), where *m* is the ports number of the interferometer, and *r* represent the amplitude. The distribution of phase is simply a uniform distribution. The statistical frequency of simulated (green bars) and experimental (red bars) amplitude and phase are shown in Fig. S16A and Fig. S16B, respectively. The amplitude shows a slight misalignment between experimental and theoretical distribution, with an overlap of ~80%. The phase distribution shows almost an excellent overlap with the theoretical prediction.

3 Phase Locking System

This section shows detailed information on the phase-locking technique, since the GBS replies on coherent superposition of photon number.

3.1 The optical design for phase locking

In our laboratory, the temperature and humidity are controlled by an air conditioning system (Vertiv Corp. Solution) in guaranteed long-term precisions of 23 ± 0.3 °C and $\pm1.5\%$, respectively. The actual temperature fluctuation is measured to be ±0.05 °C in one hours, while our measurement time is within 10 minutes. Experimentally, the drift of relative phases between the input modes can be caused by several practical noises and disturbances such like the temperature fluctuation, mechanical vibration, sound wave and so on. To stabilize the relative phase between different paths, we have developed technologies for active phase locking and passive phase stabilization.

The active phase locking is used to lock the relative phases from the first beamsplitter before PPKTP to the dichromatic mirror (DM) just in front of the interferometer. As shown in Fig. 2A in the main text, a pump laser beam is used as a reference for all the squeezed states. We take advantage of the collinearity of the PDC, that is, the pump laser shares the same propagation path as the TMSSs—both in free space and optical fibers—before entering the interferometer. After propagating through a ~2-m free space and a 20-m optical fiber, a ~10 μ W 776-nm laser is separated by a DM, which are then combined on a beam splitter with a reference laser pulse. A balanced detection scheme which is insensitive to the laser power fluctuation, is used to read out the phase information (see more details in the next subsection).

The operating principle of active locking is shown in Fig. 2A in the main text. We lock all the phases of 25 TMSSs on a reference beam by interfering the collinear 776 nm lasers with the reference laser. Note that the coherence length of laser pulse is less than 90 μ m. We have designed an optical circuit to separate one reference path into 25 different paths. The phase-locking optical circuit consists of a trapezoid piece and a rectangle piece as shown in Fig. S18, which is optically matched with our 1552-nm interferometer. In the

trapezoid piece, an input pulse is separated into 5 paths. In the same vertical plane after it, the pulses will successively accumulate a delay of τ due to the 6-mm optical length in air. As for the rectangle piece, the pulses from *a* to *e* will successively accumulate a delay of $n\tau$ due to the 6 mm optical length in SiO₂, where *n* is the refractive index of SiO₂. By combining these two pieces, one input pulse could be separated into 25 paths with different time delays which match with our rectangle and triangular interferometers for 1552-nm light. To further finely compensate the time delays, we insert many 5×5 mm optical glasses (K9) with thickness from 100 μm to 600 μm (the thickness step is 50 μ m) before entering the PBS, which guarantees high interference visibility for the convenience of phase readout.

The passive phase stabilization is employed to minimize the phase fluctuation between DM and the interferometer, while the phase inside the 3D interferometer has been demonstrated intrinsically stable (18, 38). The passive phase stabilization mounting system is shown in Fig. S17. To attenuate the mechanical vibration, first, we isolate all possible vibrations from the optical table by a honeycomb isolation plate. Second, all four interferometers, a DM and a mirror on the top of the interferometer, are bonded on an ultra-low-expansion (ULE) plate with UV-curing adhesive. The temperature of ULE plate is further controlled by a water-cooling circulator with a deviation of ± 0.02 °C. Third, All the optical elements on ULE plate are made by SiO₂ which has low coefficient of thermal expansion. Fourth, the entire optical layout is then isolated within an acrylic box to reduce the air flow.

3.2 The details of phase measurement and feedback

The phases of all paths are locked to the reference path. As shown in Fig.2C in the main text, the 776 nm laser beams and the reference laser beams are combined at the PBS, and then collected into 780HP single mode fibers. Because the collimators are optimized at 1552 nm, about 500 nW laser are couples into single-mode fiber. In order to obtain stronger signals, we employ avalanche photo diode (APD) and set the reverse bias close to the breakdown voltage. The intensity of laser from reference path and input modes are usually different, which result in a detected signal with low interference visibility. In our

experiment, we evade this problem by exploiting balanced detection scheme. As shown in Fig. 2A in the main text, we divide the collected light into two paths and send them into two APD detectors with a fiber PBS. Before the PBS, a polarization controller is used to adjust the laser power at two detectors. The differential photo current from the two APDs is amplified by a transimpedance amplifier. Another issue is that the response of the optoelectronic module will change over time. For examples, the fluctuation of the reverse bias of APDs leads to a fluctuation of signal. To make sure that we have locked the phases at a stabilized point, a heterodyne phase-locking scheme is employed. The frequency of reference laser is blue shifted by 125 kHz, which results in a phase difference of 180° between two adjacent pulses. Then if we keep the phase difference of the adjacent pulse pair stable, the relative phase will be locked to a fixed point. The signal from transimpedance amplifier is measured by an ADC chip, and then we use a STM32 microcomputer to calculate the feedback signal.

To modulate the phases of each path, we wind 5-meter-length optical fiber around a piezoelectric (PZT) ceramic cylinder which has a sensitivity of 1.5 rad/V, a response frequency of 18.3 kHz and a dynamical range of 300 rad. The diameter of PZT is selected to 6 cm to avoid photon loss in the optical fiber. In order to find the resonance frequency of the PZT, we apply alternating current to the piezoelectric ceramics and measure the currents at different frequencies under the same voltage. The measured result is plotted in Fig. S19. The current increases linearly with the frequency when the frequency is below 15 kHz. The capacitance value of piezoelectric ceramics of C = 26 pF can be calculated by formula $I = 2\pi f CU$. The current curve has a peak at 18.3 kHz and the FWHM of this peak is 0.3 kHz, which means that PZT has a resonance frequency of 18.3 kHz with a quality factor of 60.

The response of our phase modulator is measured of 1.5 rad/V. To ensure that the phase lock will not be lost during data measurement, we need a phase modulation range of 300 rad. The PID feedback signal with a voltage range of 5 V is amplified to 100 V by a non-inverting amplifier and an inverting amplifier. We apply them to the inside and outside of piezoelectric ceramics to finally achieve a 200-V modulation range.

3.3 Quantifying the phase stability

For the active phase locking, we first measure the phase stability of the 776-nm laser. These phases can be read out from the active feedback module directly. We only record the standard deviation of the input phase signal which is used for PID feedback. The measured phase with active PID control within 3.5 hours is plotted in the upper panel of Fig. 2B in the main text, the standard deviation of 776 nm laser is 0.04 rad (\sim 5 nm).

For the passive phase stabilization, we use a 1550-nm laser to measure the phase drift by a standard Mach-Zehnder configuration. By monitoring the output laser power, we can extract the relative phase between two paths. One typical example is shown in the bottom panel of Fig. 2B in the main text with a 3.5-hour measurement. The standard deviation of the passive locking part is $\lambda/180$.

Finally, we measure the phase stability of the whole system. A 1550-nm broadband white light which shares the same path with PDC photons is injected into the device, and then passes through the nonlinear crystals, optical fiber, interferometer, finally detected at the end of the fiber. For this measurement, we block 23 out of all 25 paths forming a standard Mach-Zehnder configuration to monitor the interference pattern at the output of interferometer by a spectrometer. The relative phase extracted from the interference pattern has the same stability with the phase of input TMSSs. To quantify the overall phase stability, we divide the measured phase into low-frequency and high-frequency regimes with a cutoff frequency of 0.01 Hz. The low-frequency drift is $\lambda/63$ within 1 hour and the standard deviation of high-frequency noise is $\lambda/350$.

4 Single-Photon Detection System

This section provides detailed description of the single-photon detection system for the experiment, which is composed of 100 channels of single-photon detectors, TDC and customized coincidence analyzer.

4.1 The single photon detectors

We use 7 sets of superconducting nano-wire single-photon detector (SNSPD) system with a total of 100 channels. The SNSPD devices are developed by the Shanghai Institute of Microsystems and Information Technology (39), and Quantum Opus, LLC. The devices work at ~2 K in compact cryostats, with a dark count rate of ~100 s⁻¹. The dead time of all detectors are less than 50 ns, much shorter than the pulse peirod of the pumpling laser ~4 μ s, indicating the recorded coincidence is little affected by preceding counts. The efficiencies of the 100 channels at 1552 nm, including fiber coupling loss, range from 73% to 92% with an average of 81%. Each channel is characterized and the data is listed in Table S1. In Fig. S20 we plot the ECDF of the 100 channels' efficiency.

4.2 TDC and coincidence analyzer

Once a photon triggers a count in our single-photon detector, the device outputs an electrical pulse signal which is fed into the coincidence analyzer. Our coincidence analyzer are composed of 13 8-channel FPGA boards for signal recognizing, shaping and delay tuning, and a coded central board for coincidence analyzing. For coincidence analysis, we first compensate all time delays and align all 100 channels' signals for coincidence detection. The time delay origins from two aspects: the optical path delay from the output of the interferometer to the detector, and the electrical signal delay from the detector to the coincidence analyzer. The coincidence data are streamed in real-time to a PC for further analyzing. We note that although the pulse period of our master pumping laser ($\sim 4\mu s$) is much longer than the dead time of the detectors, there can be weak leaking of the seed laser (pulse period ~ 13 ns) into the setup which would cause unwanted wrong counts. To avoid this error, the coincidence analyzer mark all such events with special tags, and these events are abandoned during further analysis of the data.

5 Experimental setup

In the last four sections, we have separately described four key parts in this experiment: squeezed light sources, 100-mode interferometer, phase locking system, and single-photon detectors. Here, we further give an overview of the whole experimental system.

A schematic diagram of our GBS experimental setup is shown in Fig. S21. First, a transform-limited pulse laser with a central wavelength of 776 nm and a repetition rate of 250 kHz is equally separated into 13 paths to pump 25 PPKTP crystals. Limited by the excitation power, two PPKTPs are cascaded and pumped with a same laser beam but with a slight cost of efficiency and indistinguishability of the second crystal. Each generated squeezed light is collected into a single-mode fiber, while most of the pump laser are filtered but leaving ~ 10 - μ W laser collected into the fiber for active phase locking. Note that this part laser is separated from the 1552-nm squeezed light by a dichromatic mirror in the front of the interferometer. To stabilize the phases of all 25 squeezed light sources, a 5-m fiber is winded around a piezoelectric cylinder which can stretch the length of the fiber to compensate the phase drift in real time. The real-time phase information is extracted from each interference pattern between a locking laser and a reference laser. To circumvent the laser intensity jitter and the response fluctuation of the APDs, balanced detection and heterodyne phase-locking schemes are employed, respectively. Then, 25 phase-locked TMSS lights are injected into a 100-mode linear optical network. Finally, the output state is detected by 100 superconducting nanowire single-photon detectors.

6 The sampling matrix

Gaussian state in the experiment is described by a two-mode squeezing operator acting on a vacuum state, which can be characterized by its covariance matrix as

$$V_{\sigma} = S_{\rm TM} V_{\rm vac} S_{\rm TM}^{\dagger} \tag{14}$$

where $V_{\text{vac}} = \frac{1}{2}I$ is the covariance matrix of vacuum state, and

$$S_{\rm TM} = \begin{bmatrix} \oplus \cosh r_k & 0 & 0 & \oplus \sinh r_k \\ 0 & \oplus \cosh r_k e^{i\phi_k} & \oplus \sinh r_k e^{i\phi_k} & 0 \\ \hline 0 & \oplus \sinh r_k & \oplus \cosh r_k & 0 \\ \oplus \sinh r_k e^{-i\phi_k} & 0 & 0 & \oplus \cosh r_k e^{-i\phi_k} \end{bmatrix}$$
(15)

The covaraince matrix of Gaussian state before and after an unitary interferometer can be related by

$$\sigma_{\rm out} = \begin{bmatrix} U & 0\\ 0 & U^* \end{bmatrix} \sigma_{\rm in} \begin{bmatrix} U^{\dagger} & 0\\ 0 & U^T \end{bmatrix}$$
(16)

When there is non-unity transmission and mode mismatch, which is the experimental case, the sampling is performed on a subspace. Therefore, we rewrite the unitary matrix to include these effects:

$$U = \begin{bmatrix} T & P \\ Q & R \end{bmatrix}$$
(17)

where T is the transform matrix between input mode and modes in the detectable subspace, while P,Q,R are the coupling between input modes with the leaked modes. Besides, we have

$$TT^{\dagger} + PP^{\dagger} = I \tag{18}$$

from the fact that $UU^{\dagger} = I$.

As the input modes corresponding to the leaked modes are in vacuum state, the output

state is

$$\begin{split} \sigma_{\text{out}} &= \begin{bmatrix} T & P & 0 & 0 \\ Q & R & 0 & 0 \\ 0 & 0 & T^* & P^* \\ 0 & 0 & Q^* & R^* \end{bmatrix} \begin{bmatrix} \sigma_{11} & 0 & \sigma_{12} & 0 \\ 0 & I/2 & 0 & 0 \\ \sigma_{21} & 0 & \sigma_{22} & 0 \\ 0 & 0 & 0 & I/2 \end{bmatrix} \begin{bmatrix} T^{\dagger} & Q^{\dagger} & 0 & 0 \\ P^{\dagger} & R^{\dagger} & 0 & 0 \\ 0 & 0 & T^T & Q^T \\ 0 & 0 & P^T & R^T \end{bmatrix} \\ &= \begin{bmatrix} T\sigma_{11}T^{\dagger} + PP^{\dagger}/2 & T\sigma_{11}Q^{\dagger} + PR^{\dagger}/2 & T\sigma_{12}T^T & T\sigma_{12}Q^T \\ Q\sigma_{11}T^{\dagger} + RP^{\dagger}/2 & Q\sigma_{11}Q^{\dagger} + RR^{\dagger}/2 & Q\sigma_{12}T^T & Q\sigma_{12}Q^T \\ T^*\sigma_{21}T^{\dagger} & T^*\sigma_{21}Q^{\dagger} & T^*\sigma_{22}T^T + P^*P^T/2 & T^*\sigma_{22}Q^T + P^*R^T/2 \\ Q^*\sigma_{21}T^{\dagger} & Q^*\sigma_{21}Q^{\dagger} & Q^*\sigma_{22}T^T + R^*P^T/2 & Q^*\sigma_{22}Q^T + R^*R^T/2 \\ \end{bmatrix} \end{split}$$

Tracing out the leaked modes, we have the covariance matrix of detectable modes

$$\sigma_{\rm out}' = \begin{bmatrix} T\sigma_{11}T^{\dagger} + PP^{\dagger}/2 & T\sigma_{12}T^{T} \\ T^{*}\sigma_{21}T^{\dagger} & T^{*}\sigma_{22}T^{T} + P^{*}P^{T}/2 \end{bmatrix}$$

$$= \frac{1}{2} \left(I - \begin{bmatrix} T & 0 \\ 0 & T^{*} \end{bmatrix} \begin{bmatrix} T^{\dagger} & 0 \\ 0 & T^{T} \end{bmatrix} \right) + \begin{bmatrix} T & 0 \\ 0 & T^{*} \end{bmatrix} \sigma_{\rm in}' \begin{bmatrix} T^{\dagger} & 0 \\ 0 & T^{T} \end{bmatrix}$$
(20)

Based on the obtained covariance matrix at the output modes, following (11) the sampling matrix is

$$O = \begin{bmatrix} 0 & I \\ I & 0 \end{bmatrix} \left(I - (I/2 + \sigma'_{\text{out}})^{-1} \right)$$
(21)

With threshold detectors the sampling distribution of non-collision events $S = \{s_i\}$ is

$$P_{\text{threshold}}(S) = \frac{\text{Tor} \begin{bmatrix} 0 & I \\ I & 0 \end{bmatrix} O_{\text{sub}}}{\sqrt{|I/2 + \sigma'_{out}|}}$$
(22)

where O_{sub} is constructed by rows S, (S + m) and columns S, (S + m) of the $2m \times 2m$ dimensional sampling matrix O(12). The function Tor is defined as

$$\operatorname{Tor}(A) = \sum_{Z \in P(\{1,2,\dots,n\})} \frac{(-1)^{n-|Z|}}{\sqrt{\det(I - A_Z)}}$$
(23)

for any matrix $A \in \mathbb{C}^{2n \times 2n}$, where $P(\{1, 2, ..., n\})$ is the set of all subsets of $\{1, 2, ..., n\}$.

7 Extracting the Phases of All TMSSs

In standard boson sampling, the relative phase between different modes does not affect the final photon distribution. However, the drift of the relative phase between different modes will totally destroy GBS, since it relies on the coherent superposition of all input squeezed states. Therefore, in our experiment, we actively lock the phases of all 25 input TMSS to a reference laser beam. Note that the phase drift of the reference laser beam doesn't affect the experiment because it is a global phase factor for the system.

A squeezing source can be described by $\xi_k = r_k e^{i\phi_k}$, where the amplitude r_k depicts amount of squeezing and the phase ϕ_k indicate the relative phase of the beam. The r_k can be obtained by measuring its intensity, i.e., count rate from photon detectors. However, the measurement of ϕ_k is more difficult. Here, we develop an experimentally feasible method through a set of small-scale GBS experiments. The running steps are as follows.

To determine the relative phases among the 25 sources, we use three sources as inputs in each run to get final 2-photon distribution. We can extract phases of the input SMSSs by fitting the final distribution since it is a function of Torontonian which is related to the phases of the input states. Specially, in this experiment, we select source 13 and 21 which are labeled as $\xi_{13} = r_{13}e^{i\phi_{13}}$ and $\xi_{21} = r_{21}e^{i\phi_{21}}$ respectively as two fixed inputs, and the third input is chosen from the remaining 23 sources ($\xi_k = r_k e^{i\phi_k}$). We set the phase of one of the source (13 and 21) to zero and measure the relative phases of other sources. With the measured r_k and transformation matrix of interferometer, in principle, the final distribution p_k uniquely depends on $\phi_{21} - \phi_{13}$ and $\phi_k - \phi_{13}$. Therefore, we can use a two-dimensional fitting to extract the values of $\phi_{21} - \phi_{13}$ and $\phi_k - \phi_{13}$ by scanning them in the range of $[0, 2\pi]$. The relative phase can be obtained when it allows a minimal total variation distance $D_k = (1/2) \sum_i |p_{\exp,k,i} - p_{\operatorname{cal},k,i}|$. A typical two-photon total variation distance is plotted in Fig. S22 which shows evident peak and valley.

We use this method to calibrate the relative phases of the all 23 squeezing sources, the result is shown in Fig. S23. The extracted relative phases between sources 13 and 21 throughout all 23 measurements remain stable with a mean of 1.18 and standard deviation of 0.06, which sufficiently demonstrate the correctness of our method.

As a comparison experiment, we perform the same small-scale GBS without phase locking and letting the phase free running. The fitting results is plotted in Fig. S24 which clearly shows that the relative phase fluctuates as function of the time if without active control.

8 Experimental validation of GBS

Here we present detailed description of developed methods to give sufficient evidences to show that the output samples are correctly drawn from the true quantum GBS machine rather than other classically simulable models.

8.1 Comparison of photon distribution with possible hypotheses

By the nature of TMSS, the output photon number of GBS is uncertain and shows a Poisson-like distribution, as shown in Fig. 3C and 3E (red dot) in the main text. This distribution results from intrinsic quantum interference of 25 all TMSSs. With classical light input, however, the detected photon number may be totally different since there is no (or much less) interference among input states. This inspires us to explore the output photon distribution with different input states, thereby revealing the quantum nature of GBS and naturally exclude classical mockups.

There are several plausible models with classical input should be focused on. The first one is the thermal distribution—caused by the excessive loss in the system. The second one is the distinguishable SMSS distribution—resulting from the spatial or temporal mismatch of the input photons. Note that the sampling with classical input can be efficiently solved by conventional computers.

We first consider sampling from an 2K-mode input thermal state $\rho = \bigotimes_{i=1}^{2K} \rho_i^{\text{th}}$. We choose the i, K + i thermal state with the mean photon number $\bar{n}_i = \bar{n}_{K+i} = \sinh^2 r_i$, which is equal to the mean photon number of one mode of the *i*-th TMSSs. The thermal state can be regarded as a Gaussian statistical mixture of the coherent state $\hat{\rho}_i^{\text{th}} = \int P_i^{\text{th}}(\alpha) |\alpha\rangle \langle \alpha | d^2 \alpha$ according to the Glauber-Sudarshan P function, where $P_i^{\text{th}}(\alpha)$ is:

$$P_i^{\rm th}(\alpha) = \frac{1}{\pi \langle \bar{n}_i \rangle} \exp\left(-|\alpha|^2 / \langle \bar{n}_i \rangle\right)$$

Now we show how to efficiently sampling from GBS with thermal input (40). First choosing a random set as input coherent states with amplitudes $\{\alpha_i\}_{i=1}^{2K}$ from the probability distributions $\{P_i^{\text{th}}(\alpha)\}_{i=1}^{2K}$. When the multimode coherent state $\{\alpha_i\}_{i=1}^{2K}$ passes through the linear optical network U, the output is a new multimode coherent state, and the amplitude of the *j*-th mode is

$$\beta_j = \sum_i \alpha_i U_{ij}.$$

Considering the threshold detection, the probability to have a photon click at output port j-th is

$$1 - p_j(0) = 1 - e^{-|\beta_j|^2},$$

thereby, we can generate many samples efficiently. The simulated photon distribution is presented in Fig. 4E (blue dots).

Another model we want to test is distinguishable SMSS states. The input state is $\rho = \bigotimes_i |r_i\rangle \langle r_i|$. Since distinguishable squeezed states do not interfere with each other, we just sampling from *i*-th SMSS to generate an output $s_i = (x_{i1}, x_{i2}, ..., x_{im})$ independently, then the classical mixture $s = \sum_i s_i$ is a sample of GBS with all SMSSs input together. Sampling from GBS with one distinguishable input SMSS state $|r_i\rangle \langle r_i|$ can be efficiently done as follow: First expand state at Fock-state basis,

$$|r_i\rangle = \frac{1}{\sqrt{\cosh r_i}} \sum_{n=0}^{\infty} \frac{\sqrt{(2n)!}}{2^n n!} \tanh r_i^n |2n\rangle = \sum_{n=0}^{\infty} c_n |2n\rangle$$

then random choose a photon number 2n from probability distribution $\{|c_n|^2\}$, finally sample from GBS with Fock-state $|2n\rangle$ input, all the steps above can be efficiently simulated by classical computers.

8.2 Sample density distribution and heavy output generation ration test

In the sparse regime of Gaussian boson sampling, the frequency/probability distribution cannot be efficiently reconstructed by the experimental samples due to the exponential size of sample space. Here, we develop intuitive visualization and computation-friendly statistical tools to tame the validation of sparse sampling. We start to describe the method to visualize the experimental sample density distribution (see Fig. 3H, gray line). It allows us to directly judge the samples are non-uniform and also allows us to reconstruct the approximate probability distribution of the samples.

The basic process includes two steps. (1) Generating large-N uniform random samples, computing their probabilities, and sorting the probabilities. In this step, we obtain a reference curve of the theoretical probabilities by plotting the sorted probabilities of random samples. (2) Computing the probabilities of experimental samples, comparing the probabilities to the reference curve, and putting marking points on the curve with the same probability values. We can further insert vertical lines from the marking points to the x axis to enhance the visual effect. In this step, we obtain the density distribution of sparse experimental samples.

From the sample density distribution (see Fig. 3H), we can directly see the experimental samples more likely to shot the curve at the regime of heavy probabilities. It is an intuitive signal of non-uniform distribution. This observation further inspires us to design a heavy output generation (HOG) ratio test to distinguish the experimental samples from other mock-up samples. When bosons go through a random network, the output patterns with heavy probabilities are due to the constructive quantum interference of the possible paths. It is hard for a classical mockup sampler to identify the right patterns of constructive interference, therefore, a mockup sampler will not likely generate samples at the output patterns with heavy probabilities. Given N experimental samples and N mockup samples, we calculate the likelihood of these samples coming from the ideal theoretical distribution, as $Pr_{ideal}(Samples_{mockup})$, respectively. We use the likelihood ratio r_{HOG} as the indicator to account the heavy output generation:

$$r_{\rm HOG} = \frac{{\rm Pr}_{\rm ideal}({\rm Samples}_{\rm exp})}{{\rm Pr}_{\rm ideal}({\rm Samples}_{\rm exp}) + {\rm Pr}_{\rm ideal}({\rm Samples}_{\rm mockup})}$$

By increasing the number of samples, if the likelihood ratio approaches 1, the experimental samples have an overwhelmingly possibility coming from the ideal distribution over the mockup samples. Our experimental HOG analyses for photon clicks from 34 to 38 on Sunway TaihuLight is shown in Fig. 3G in the main text, and the other results 26 to 33 for photon clicks from 26 to 33 is shown in Fig. S26.

A traditional statistical test in the previous experiments is the Bayesian test. It is used to argue the experimental samples are more likely generated from ideal distribution than the mockup distribution. With 1:1 prior probabilities of the two distributions, the confidence of experimental samples from the ideal distribution is

$$c_{\mathrm{Bayesian}} = \frac{\mathrm{Pr}_{\mathrm{ideal}}(\text{Samples}_{\mathrm{exp}})}{\mathrm{Pr}_{\mathrm{ideal}}(\text{Samples}_{\mathrm{exp}}) + \mathrm{Pr}_{\mathrm{mockup}}(\text{Samples}_{\mathrm{exp}})}$$

Although the two formulas are similar, there are several advantages of the HOG ratio test comparing to the Bayesian: (1) HOG ratio test has an intuitive physical meaning for the constructive quantum interference. (2) HOG ratio test only requires the mockup sampler to generate samples, so the mockup sampler does not need to have the ability to model the generating probabilities. (3) HOG is computation-friendly. HOG only involves the probabilities of one distribution, so the calculation of relative probabilities between samples is enough to accomplish the test, while the Bayesian needs to calculate the absolute probabilities of the two distributions.

9 Benchmarking GBS with Sunway TaihuLight supercomputer

We design and implement a high-precision and high-performance classical algorithm on the state-of-the-art supercomputer, Sunway TaihuLight, to establish the quantum computational supremacy frontier of Gaussian boson sampling. The Sunway Taihulight system has 40960 computing nodes, where each node has four core groups with total 3 TFLOPS performance. The whole system has a peak performance of 125 PFLOPS.

The task is to calculate the Torontonian function. The matrix function is defined as

Tor(A) =
$$\sum_{Z \in P_N} (-1)^{N-|Z|} \frac{1}{\sqrt{|\det(I - A_Z)|}},$$

where A is a $2N \times 2N$ input matrix, A_Z is a submatrix by selecting the k-th and (N+k)-th row and column of matrix A with $k \in Z$, and P_N is the power set of $\{1, 2, ..., N\}$. There are 2^N terms in the summation and the computational complexity is $O(N^3 2^N)$.

To scale the simulation to $N \ge 50$, we use 256-bit precision in the calculation based on an architecture-specific instruction set and we use an advanced partition for massive parallel to optimize the cache-level storage and the loading balance. We implement the highest sustained performance of 2.78 PFLOPS. The execution time for N = 50 is 170891 seconds (about 2 days).



Supplementary Figure S1 | **An overview of our GBS implementation.** The GBS device is realized by sending 50 single-mode squeezed states (SMSSs) into a 100-mode interferometer, and then sampling from the output distribution using 100 single-photon detectors.



Supplementary Figure S2 | **The pump laser system in our experiment. a**, schematic of the connection between the subsystems, we use two Verdi laser to respectively pump the mode-locked Ti:Sapphire oscillator (Mira 900) and the femtosecond Ti:Sapphire amplifier (RegA 9000). External spectrum shaping and Expander/compressor optical layouts are designed to improve the quality of output pulse. b, optical setup for spectrum shaping. We use two near-infrared grating to diffract and combine the seed laser beam and adaptive deformable mirrors to modulate the beam. The slit is inserted to cut down the sideband which make the laser share a symmetrical spectral intensity distribution. Optical 4f-system is used for collimated light beam.



Supplementary Figure S3 | **The reconstruction spectral intensity and phase of pump laser. a**, the spectral intensity distribution of pump laser therein red Gaussian curve is fitted to guide the eyes. **b**, the spectral phase of pump laser reconstructed from the measurement of FROG.



Supplementary Figure S4 | **Design of the PPKTP crystals.** The blue line shows the poling function along the crystal used in this experiment, resulting in the normalized field amplitude shown as red circles, which is consistent with an ideal Gauss error function (the green line).



Supplementary Figure S5 | **Optical setup of our squeezing sources.** We use an array of beam splitters to averagely separate the pump laser beam. Then, the laser beams are guided to pump each sources which are installed in an array of temperature-controlled box of TEC. After each source, we use a dichroic mirror to reflect the down-conversion squeezing photons and couple them into a single-mode fiber. The details of source are shown in the main text. PBS: polarizing beam splitter, DM: dichroic mirror, PPKTP: periodically poled KTP crystals.



Supplementary Figure S6 | The photon pair's wavelength dependence on the temperature of the PPKTP crystal, indicating a good approximation of linear dependence around the degenerate central wavelength. In experiment the degeneracy is tuned by PID temperature control and the wavelength detuning is controlled to be less than 1 nm (> 99% indistinguishability).



Supplementary Figure S7 | The measured joint spectrum of the photon pair with PPKTP tuned at central-wavelength-degenerate temperature, after 12nm filtering, indicating the two photons are free of frequency correlations. The spectral purity after filtering is 0.99.



Supplementary Figure S8 | (A), (B) The spectrum of H(V) polarization mode of the photon pair, each corresponds to a central wavelength of 1552nm and a FWHM of 12.4(13.2) nm before filtering.(C), (D) The spectrum of H(V) polarization mode of the photon pair after 12-nm filtering.



Supplementary Figure S9 | (A) The curve shows the dependence of the measured $g^{(2)}(0)$ on the detection probability of the threshold detectors, with fixed purity $\mathbb{P} = 0.95$. The dashed line shows the same measurement result with an ideal detector. (B) An experimentally measured $g^{(2)}(0)$ histogram. The bar on the left is the zero-time-delay coincidence counts while the central (right) bar is the coincidence counts with 1(2) pulse period delay.



Supplementary Figure S10 | Illustration of our hybrid encoding photonic circuit. Our experiment exploit spatial and polarization degrees of freedom to encoding our multimode photonic circuit. In the first stage, H-polarized photons and V-polarized photons interfere in a 3 dimensional interferometer respectively. In the second stage, polarization interferences are realized by an array of Mach-Zehnder-type interferometers which make our circuit fully connected.



Supplementary Figure S11 | Schematic design for interferometer. (A) and (B) represent the cross-sectional view of our retangular and triangular pieces respectively. (C) Our three dimensional interferometer using retangular and triangular pieces as building blocks. The 25 TMSSs are firstly aligned in a 5*5 array and injected into triangular piece. Then, after passing through the retangular piece, the 50 spatial output modes are distributed into two 5*5 arrays.



Supplementary Figure S12 | Path delay between different output modes. Using white light interferometer, we test the optical path delays of different input combinations. The standard deviation is less than 3 μm from our sample testing.



Supplementary Figure S13 | **The estimated photon indistinguishability after path delay.** By taking into account the path delays between the different input-output combinations, the photon indistinguishability is estimated. The subplot is plotted to show the coherence length of the TMSS source.



Supplementary Figure S14 | Schematic diagram (A) and photograph (B) of our photonic network. The experimental setup is built on an optical table with an area about 3 square meters. In the input optics region, 25 TMSSs are injected into the photonic network. Correspondingly, at lower right, 25 phase-locking light is collected. The output modes of our photonic network are separated to 100 spatial modes by using mini-mirrors and PBSs.



Supplementary Figure S15 | Unitary test of the reconstructed matrix. The real and imaginary parts of the UU^{\dagger} are shown in A and B, respectively.



Supplementary Figure S16 | Randomness of the reconstructed matrix. A, the normalized statistical frequency of all 5000 amplitude elements of the reconstructed matrix (green bars). The red histogram is from 1000 simulated unitary matrices according to Haar measure, and red solid line is the theoretical fitting. The calculated overlap is 80%, indicating a good agreement between experiment and Haar measure. B, both the experimental and simulated phase elements shows a uniform distribution from $-\pi$ to π —an excellent match between experiment and Haar measure.



Supplementary Figure S17 | The design for passive phase stabilization mounting system. At the bottom, a honeycomb-structured vibration-isolation plate is placed on the optical platform, for further attenuating mechanical vibration from the ground. On top of the plate, we installed a liquid-cooling aluminum board to maintain a temperature stability of ± 0.02 °C. A piece of 20mm-thick ultra-low expansion glass (ULE) is adhered to the liquid-cooling board, with the contact area sealed by silicone grease. The ULE's upper surface functions as the the ultimate 200 mm*200 mm platform for gluing and fixing the interferometer.



Supplementary Figure S18 | The phase-locking optics. A, a trapezoid-like interferometer separate one input beam into 5 paths denoted as $1, 2, \dots, 5$. B, a rectangular interferometer further separate one beam (for example path 1) into 5 paths denoted as a, b, \dots , e. C, a reference input beam propagating through two cascaded interferometers result in 25 beams for active phase locking.



Supplementary Figure S19 | **Effective current at different frequencies**. We apply signals of a same voltage and different frequencies to piezoelectric ceramics to measure the resonance frequency of piezoelectric ceramics. The resonance frequency is 18.3 kHz with a quality factor of 60.



Supplementary Figure S20 | **Empirical cumulative distribution function (ECDF) of detector efficiency.** The 100 channels' efficiency are characterized and displayed in ECDF. The average efficiency is 0.81 with a maximum of 0.92 and a minimum of 0.73.



Supplementary Figure S21 | A schematic diagram of GBS experimental setup. The system can be devided into four parts: squeezed light sources, phase locking system, interferometer and single-photon detectors. First, a transform-limited pulse laser with an overall power of 1.4 W are equally divided into 13 paths to pump 25 independent PPKTP crystals. The generated collinear two-mode squeezed light in each path is collected into a single-mode fiber. To actively stabilize the phase of each squeezed light source, a 5-m fiber is winded around a piezo-electric (PZT) cylinder to compensate the phase change so that we can stabilize the phase in a fixed point with PID feedback control. Then, 25 phase-locked TMSSs lights are injected into a 100-mode interferometer to talk with each other. Finally, the output state are detected by 100 superconducting nanowire single-photon detectors.



Supplementary Figure S22 | Typical two-photon total variation distance in our phase fitting tests. We use two-dimensional fitting method to estimate the relative phases between our three input sources. The X and Y axises represent the relative phases and the Z axis is the total variation distance between the experimental two-photon distribution and calculated two-photon distribution.



Supplementary Figure S23 | **The relative phases of our 25 sources.** Three sources including two fixed sources (source 13 and source 21) and another to-be-measured source are used as input in each experiments. The blue points show the fitted relative phases of each sources and the red points represent the relative phase between source 21 and 13 during these experiments.



Supplementary Figure S24 | Total variation distance (TVD) in our phase fitting test without and with active feedback. Phase fitting test to observe the phase fluctuation with (A) and without (B) active feedback. The color shows the value of TVD between experimental and theoretical two-click distribution. The diagram series of TVD with active feedback is stable, but changes randomly without active feedback.



Supplementary Figure S25 | **HOG ratio test and Bayes test. A**, The heavy output generation (HOG) ratio test. The experimental samples from quantum device and the mockup samples from hypothesis distribution are compared to determine which one has a higher likelihood from the ideal theoretical distribution. B, The traditional used Bayesian test. The likelihood of the experimental samples from the ideal theoretical distribution and the mockup hypothesis distribution are compared.



Supplementary Figure S26 | HOG analyses of photon clicks from 26 to 33. The red lines are from the HOG test on our experimental samples, while the blue line are from thermal distributions. The stark differences of fast convergence to 1 and 0 for GBS experiments and thermal samples indicates that our quantum machine is far from thermal sampler.



Supplementary Figure S27 | Heavy output generation of samples for the number of output clicks ranging from 26 to 29.



Supplementary Figure S28 | The time and cost for classical simulation on a supercomputer. The execution time of one Torontonian function on Sunway TaihuLight with input matrix of size up to N = 50 (the blue points). In the regime of N > 30, the Sunway system starts to fully load and the scaling is linear in the logarithmic time. We extrapolate the execution time to N=80 (the red points), where the 256-bit precision in the benchmarking keeps sufficiently precise. The standard cost of the Sunway system is USD 0.03 for one core group per hour. The total in-principle cost to calculate one Torontonian function is shown in the right axis.

| No. | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
|------------|------|------|------|------|------|------|------|------|------|------|
| Efficiency | 0.87 | 0.84 | 0.84 | 0.83 | 0.83 | 0.81 | 0.81 | 0.78 | 0.73 | 0.84 |
| No. | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 | 19 | 20 |
| Efficiency | 0.83 | 0.83 | 0.83 | 0.82 | 0.81 | 0.81 | 0.81 | 0.80 | 0.80 | 0.80 |
| No. | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 | 29 | 30 |
| Efficiency | 0.86 | 0.82 | 0.82 | 0.81 | 0.80 | 0.80 | 0.80 | 0.79 | 0.79 | 0.79 |
| No. | 31 | 32 | 33 | 34 | 35 | 36 | 37 | 38 | 39 | 40 |
| Efficiency | 0.89 | 0.88 | 0.86 | 0.82 | 0.92 | 0.92 | 0.88 | 0.86 | 0.86 | 0.84 |
| No. | 41 | 42 | 43 | 44 | 45 | 46 | 47 | 48 | 49 | 50 |
| Efficiency | 0.80 | 0.81 | 0.79 | 0.80 | 0.80 | 0.80 | 0.80 | 0.85 | 0.82 | 0.80 |
| No. | 51 | 52 | 53 | 54 | 55 | 56 | 57 | 58 | 59 | 60 |
| Efficiency | 0.82 | 0.83 | 0.81 | 0.80 | 0.79 | 0.79 | 0.81 | 0.81 | 0.79 | 0.81 |
| No. | 61 | 62 | 63 | 64 | 65 | 66 | 67 | 68 | 69 | 70 |
| Efficiency | 0.80 | 0.83 | 0.76 | 0.83 | 0.82 | 0.79 | 0.80 | 0.80 | 0.77 | 0.77 |
| No. | 71 | 72 | 73 | 74 | 75 | 76 | 77 | 78 | 79 | 80 |
| Efficiency | 0.87 | 0.86 | 0.86 | 0.84 | 0.83 | 0.83 | 0.82 | 0.82 | 0.82 | 0.82 |
| No. | 81 | 82 | 83 | 84 | 85 | 86 | 87 | 88 | 89 | 90 |
| Efficiency | 0.82 | 0.82 | 0.82 | 0.81 | 0.81 | 0.81 | 0.81 | 0.81 | 0.81 | 0.80 |
| No. | 91 | 92 | 93 | 94 | 95 | 96 | 97 | 98 | 99 | 100 |
| Efficiency | 0.80 | 0.80 | 0.80 | 0.79 | 0.79 | 0.79 | 0.79 | 0.77 | 0.77 | 0.77 |

 Table S1 | Efficiencies of the 100 channels of single photon detectors.

| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 15 36 7 36 87 97 87 </th |
|---|---|--|---|
| | $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | $\begin{array}{cccccccccccccccccccccccccccccccccccc$ | 29 40031 0013 40043 40123 00256 00375 00375 00272 00181 00303 00232 00181 00303 00395 00375 003 |
| $ \begin{array}{cccccccccccccccccccccccccccccccccccc$ | 0.000 0.0000 </td <td>0 mm 0.000</td> <td>0 0</td> | 0 mm 0.000 | 0 |

Table S2 | Real part of the reconstructed matrix of the 100-mode interferometer.

| 44-a 0012 0072 0073 00191 0.016 0022 0.0073 0.021 0.010 0.055 0.023 0.015 0.0073 0.023 0.007 0.023 0.003 0.008 0.007 0.023 0.007 0.003 0.0 | 1 1 0 0 0 0 0 |
|--|---|
| | 1 1 |

Table S3 \mid Image part of the reconstructed matrix of the 100-mode interferometer.

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