
Supplementary information

Continuous-variable multipartite entanglement in an integrated microcomb

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Supplementary Information: Continuous-Variable Multipartite Entanglement in an Integrated Microcomb

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1 Continuous-variable entanglement microcomb chip and experimental setup

The continuous-variable multi-qumode entanglement microcomb chips were fabricated on an 800nm-thick low-loss silicon nitride platform in LIGENTECH. Aiming for an efficient four-wave-mixing (FWM) in the microring resonator, it requires a small anomalous group-velocity dispersion (GVD) at the wavelength of nearly 1550 nm. This can be achieved by tailoring the structural dispersion of waveguides by optimizing its geometrical parameters. The simulation of structural dispersion is shown in Fig.S1b. The waveguide width of 1000 nm was chosen to provide a small anomalous dispersion. The cross section of waveguides was chosen as 1000 nm × 800 nm, and the mode field distribution for the TE fundamental mode is shown in Fig.S1a. The propagation loss of silicon nitride waveguides was measured to be around 0.1dB/cm. An intrinsic quality factor of 1 million was measured. The microring was devised with an FSR of $\Delta f_{\text{cavity}} = 200$ GHz (199.9 GHz FSR was measured in the experiment). Polychromatic bright laser beams (in our experiment, we used three beams) at the resonant wavelengths of the microring, that is $\lambda(P_{-3}) = 1545.475$ nm (194.115THz), $\lambda(P_0) = 1550.245$ nm (193.518THz), and $\lambda(P_3) = 1555.041$ nm (192.921THz), were used to synchronously excite the FWM processes in the microring (see Fig.2e and Fig.2f). The power of three pumps was chosen as: $I(P_{-3}) = 123$ mW, $I(P_0) = 242$ mW, and $I(P_3) = 119$ mW, by which the microring was operated below the threshold of optical parametric oscillation (OPO) for generating the eight-qumode Gaussian state. The three pump beams were coherently (phase-locked) prepared by using an EO comb system with a mode spacing of $\Delta f_{\text{EO}} = 39.98$ GHz, which is 1/5 of the microring FSR.

Figure 2g shows the experimental setup. The EO comb was prepared in the following way: a CW laser (OEwave) at $\lambda(P_0)$ wavelength was first amplified by an EDFA to 200 mW, and then modulated by three cascaded electro-optic phase modulators (EOMs, Eospace) at a frequency of 39.98 GHz. The EOMs were driven by amplified electrical signals from an RF signal generator (Rohde Schwarz, SMA100B), which were synchronized by electrical phase shifters (EPSs). The output power of the electrical amplifiers (AMPs) was about 35 dBm. The generated EO comb was then sent to a programmable line-by-line wave-shaper (Finisar 16000A), which allows arbitrary controls of the intensity and phase for each EO comb line. The multi-pump beams were selected by the wave-shaper and amplified by another EDFA. Before injection into the quantum microcomb chip, the amplified spontaneous emission (ASE) noises near the signal and idler frequencies were cleaned up using a wavelength-division multiplexing (WDM) filter (not shown in Fig.2g) with a channeling spacing of 200 GHz, matching with that of the microcomb. The polarization state of the multi-pump beams was optimized to excite the TE mode in the waveguide (see simulated mode distribution in Fig.S1a). The quantum microcomb chip was packaged on a temperature-stabilized stage (see photograph in Fig.2a). The below-threshold eight-qumode Gaussian states were generated in the microcomb using the tri-chromatic synchronously pumping. The input of pump light and output of entangled light were accessed using lensed single-mode fibers. The resonant frequencies of the microcomb can be thermal-optically tuned using an integrated microheater fabricated on the top of the silicon nitride microring (see image in Fig.2b). The resonances of microcomb were actively stabilized and locked to the $\lambda(P_0)$ wavelength of the pump laser using the side-of-fringe locking technique. In the channel of signal beams, the residual pumps (excepted at λ_{+3} frequency) were well removed from the generated entangled light using cascaded DWDM filters.

We adopted a method named as coherent polychromatic pumping-detection (CPPD) for the characterization of the generated multipartite entanglement states, as shown in Fig.2g and Fig.S3. The CPPD is a combination of methods of “multi-pump” for pumping and “operating a base change via a LO” for detection, which is a powerful way to generate, manipulate and characterize frequency-domain cluster-type states [1–3]. In our work, we adopted these two advanced technologies and combined them coherently as we named as CPPD, which allows for globally phase-locked throughout the polychromatic pumping and detection for the characterization of cluster-type states. Note that this globally phase-locked CPPD is essential to our experiment to fully characterize the covariance matrix with q - p quadrature correlations. The covariance matrix of the generated Gaussian states in the third-order microcomb exhibits strong q - p quadrature correlations (see Section 2.4), necessitating a comprehensive phase-locking approach by which the relative phase of every qumode with each other and with its corresponding LO must be stabilized. In the cases of path or time-domain cluster-type states, probe beams are usually used to individually lock the phases between squeezed modes [1, 2, 4, 5]. Our CPPD method employs one of the three pumps as a phase reference (in our experiment, we used the f_{+3} beam as the reference). Here is more details for the global phase locking in CPPD. First of all, phases between multiple pump beams from the same EO comb are intrinsically stable. As the multi-pump beams propagate with multimode squeezed beams along the same optical fiber, from the excitation, to on-chip microcomb, and to detection (as depicted by the bottom path in Fig.S3c), the relative phases between the pump and squeezed beams are stable. Similarly, the polychromatic LO beams and the reference beam (f_{+3} beam) also co-propagate along the same fiber (as depicted by the top path in Fig.S3c), enabling the preparation of reliable LOs and stabilizing the relative phase between LO and f_{+3} beams. Subsequently, the reference f_{+3} beam, extracted after beam splitter (the beam splitter in the homodyne detection closes the interferometer), is recycled to achieve a stable overall PID-locked phase between LOs and

multimode squeezed beams. This CPPD method results in a system where only the overall phase between the LO and squeezed beams remains unfixed. Using a single PID locking, we achieved the phase locking of eight-mode states with LOs (this method can be scaled up to large number of modes). That means, among the eight frequency modes, a robust reference framework is established for the covariance matrix reconstruction. Furthermore, leveraging a wave shaper, we can access any eigen-supermodes and nullifiers of the entangled states. By combing the advanced techniques of “multi-pump” and “operating a base change via a LO”, the CPPD method streamlines the system’s complexity for full generation, manipulation and measurement of multimode entanglement in the microcomb. Without such method, it represents technological challenging for the demonstration of first integrated-optical multipartite entangled states. In addition, this method enabling a fully phase stable system throughout the generation, manipulation and measurement of entangled states, could provide a robust foundation for measurement-based quantum computing that basically requires precise and phase stable preparation, control, and detection of every qumodes. Finally, in future, the devices of EO comb, wavelength multiplexing, beam splitter, and photodiodes potentially can be monolithically integrated on a single chip, allowing for scalable and controllable quantum photonic devices for quantum computing and information processing.

Figure S3 shows three schematic diagrams for monochromatic, bichromatic, and polychromatic homodyne detections. The LO beams interfere with the multimode Gaussian beams on a balanced beamsplitter, and quadrature correlations between different modes can be extracted from the photon current difference. The photodiodes (PDs) have a quantum efficiency of 95%. Monochromatic homodyne detections were implemented to calibrate the short noise levels in each mode. Bichromatic homodyne detections were implemented to characterize the covariance matrices in the quantum frequency comb-modes. Polychromatic homodyne detections were implemented to characterize and verify the multipartite entanglement of supermodes. A flexible switching of LOs was enabled by operating the wave-shaper. All measurements in the experiment were performed with the LO optical power of 10 dBm, well above the detector noise level. The characterization of local oscillator noises is provided in Fig.S2. The characterizations of quadrature squeezing and multipartite entanglement were implemented at the 5-500 MHz sideband frequency of the pump. In the regime of 5-500 MHz, the common mode rejection ratio of > 10 dB was measured. At the lower frequency, below 3 MHz, there is still a lot of noise. In most of our measurements, we choose 5 MHz as the basic sideband frequency to accurately characterize squeezing and entanglement, at which near optimal measurements can be implemented (see discussions in Sec.2.3). The system loss after the microcomb was measured to be $\eta = 0.473$ (escape efficiency is not included), including the coupling loss between chip and fiber, loss in off-chip optical components, and detection loss.

2 Generation, manipulation and measurement of continuous-variable multi-qumode entanglement

2.1 The procedure for the generation, manipulation, and measurement of various supermode multipartite entanglement with cluster-type structures and vanishing off-diagonal nullifier correlations

We employ a building-up approach that utilizes linear operations, based on the ref.[6], to create various supermode multipartite entanglement with cluster-type structures. This methodology is illustrated in Fig.S5, which shows the procedure for the generation of these entangled states in the microcomb. For the sake of simplicity, the subsequent discussion disregards the impact of coupling loss. We firstly apply the Williamson decomposition to the measured covariance matrix of frequency comb-modes:

$$\Sigma(\hat{q}, \hat{p}) = SNS^T. \quad (S1)$$

We obtain the symplectic operation S generated by the multi-pumping nonlinear process and the thermal components N arising from Bragg scattering between the relevant frequency modes and others (where N is nearly an identity matrix in our experiment, with the maximum term being 1.01). Then we apply the Bloch-Messiah decomposition to obtain the eigen-supermodes:

$$S = O_1 K O_2. \quad (S2)$$

The orthogonal symplectic operation O_1^T is applied on the frequency modes to get squeezing and anti-squeezing of eigen-supermodes in our experiment. We define the quadrature of eigen-supermodes as $(\hat{q}_e, \hat{p}_e)^T = O_1^T(\hat{q}, \hat{p})^T$, whose covariance matrix is:

$$\Sigma(\hat{q}_e, \hat{p}_e) = O_1^T \Sigma(\hat{q}, \hat{p}) O_1 = K O_2 N O_2^T K. \quad (S3)$$

The covariance matrix of eigen-supermodes is actually diagonally dominant as shown in the measured results in Fig.3d in main text, owing to the low thermal component. For theoretical simplicity, the covariance matrix can be approximately written as:

$$\Sigma(\hat{q}_e, \hat{p}_e) = K^2 = \begin{pmatrix} \text{diag}(e^{R_l}) & 0 \\ 0 & \text{diag}(e^{-R_l}) \end{pmatrix}. \quad (S4)$$

Crucially, it is essential to finely adjust the squeezing strength of the eigen-supermodes (represented as e^{-R_l} for l -th eigen-supermode), and subsequently apply additional linear operations to generate well-approximated supermode cluster-type states within the finite squeezing region [6]. In our experiment, we introduce a new constraint condition to the multimode linear operations, building upon the ref. [6], and determine the associated squeezing strengths to force the off-diagonal nullifier correlations into vanishing. The symplectic operation T_1 corresponding to the linear unitary operation U is written as:

$$T_1 = \begin{pmatrix} \text{Re}(U) & -\text{Im}(U) \\ \text{Im}(U) & \text{Re}(U) \end{pmatrix}, \quad (S5)$$

where $\text{Re}(U)$ and $\text{Im}(U)$ denote the real and imaginary part of unitary matrix U , respectively. To ensure the elimination of divergent noise terms caused by antisqueezing, the ref.[6] has established the following condition:

$$\text{Im}(U) - A \cdot \text{Re}(U) = 0, \quad (S6)$$

where A is the adjacent matrix of expected cluster-type states. However, this condition only ensures that the prepared states asymptotically can approach the cluster-type states in the infinite squeezing region; it cannot force the off-diagonal nullifier correlations into vanishing for the approximate cluster-type states, when physical finite squeezing is available in experiment (see Supplementary Section 3.3). According to the unitary condition on U ($Re(U)Re(U)^T + Im(U)Im(U)^T = I$, $Re(U)Im(U)^T = Im(U)Re(U)^T$), Eq.S6 is equivalent to:

$$Re(U)Re(U)^T = (I + A^2)^{-1}, \quad (S7)$$

which includes $n(n+1)/2$ constraint equations for n -dimensional $Re(U)$. To uniquely determine $Re(U)$ and then linear operation U , the further condition should be taken.

We apply the undetermined symplectic operation T_1 on the eigen-supermodes to obtain the transformed supermodes for the approximate cluster-type states with finite squeezing: $(\hat{q}_c, \hat{p}_c)^T = T_1(\hat{q}_e, \hat{p}_e)^T = T_1 O_1^T(\hat{q}, \hat{p})$. The correlation matrix of nullifiers composed by quadrature $(\hat{q}_c, \hat{p}_c)$ can be derived:

$$\begin{aligned} cov[\hat{p}_c - A \cdot \hat{q}_c] &= (-A \quad I) T_1 K^2 T_1^T \begin{pmatrix} -A \\ I \end{pmatrix} = (Re(U) + A \cdot Im(U)) \cdot diag(e^{-R_l}) \cdot (Re(U) + A \cdot Im(U))^T \\ &= (I + A^2) Re(U) \cdot diag(e^{-R_l}) \cdot Re(U)^T (I + A^2) \\ &= (Re(U)^{-1})^T Re(U)^{-1} \cdot \{Re(U) diag(e^{-R_l}) Re(U)^T\} \cdot (Re(U)^{-1})^T Re(U)^{-1} \\ &= (Re(U)^{-1})^T \cdot diag(e^{-R_l}) \cdot Re(U)^{-1} \end{aligned} \quad (S8)$$

A straightforward approach can be taken to force the off-diagonal nullifier correlations into vanishing is the following: the column vectors of $Re(U)^{-1}$ are weightedly orthogonal to each other with the weight matrix $diag(e^{-R_l})$. A possible construction is to force the column vectors of $Re(U)$ orthogonal to each other, giving another $n(n-1)/2$ constraint equations to uniquely determine $Re(U)$ and $Im(U)$ (Note that no limitation is imposed on the norm of column vectors of $Re(U)$). This leads to:

$$Re(U)^T Re(U) = D, \quad (S9)$$

where D is a diagonal matrix determined by $Re(U)$. In experiment, we can adjust the polychromatic pump configurations to make the squeezing strength (the weight matrix for $Re(U)^{-1}$) proportional to D :

$$diag(e^{-R_l}) = \lambda D, \quad (S10)$$

where λ is a constant value. From Eq.S9, we can derive that:

$$cov[\hat{p}_c - A \cdot \hat{q}_c] = \lambda (Re(U)^{-1})^T \cdot D \cdot Re(U)^{-1} = \lambda I. \quad (S11)$$

This implies that by finely tuning the pump configurations to approximately satisfy the Eq.S10, the correlation matrix of nullifiers would be nearly a identity matrix multiplied by a constant λ . This provides an experimentally feasible method to prepare multipartite entanglement with cluster-type structures in the microcomb. The calculated $Re(U)$ s for various cluster-type structures are shown below:

$$Re(U) = \begin{pmatrix} -0.0598 & 0.2699 & -0.0367 & -0.7227 \\ 0.4366 & 0.0967 & 0.4466 & -0.0227 \\ -0.0967 & 0.4366 & 0.0227 & 0.4466 \\ 0.2699 & 0.0598 & -0.7227 & 0.0367 \end{pmatrix}, \text{ four-mode linear-type} \quad (S12)$$

$$Re(U) = \begin{pmatrix} -0.3101 & -0.0621 & -0.3142 & -0.6335 \\ -0.0621 & 0.3101 & 0.6335 & -0.3142 \\ -0.3101 & -0.0621 & 0.3142 & 0.6335 \\ -0.0621 & 0.3101 & -0.6335 & 0.3142 \end{pmatrix}, \text{ four-mode box-type} \quad (S13)$$

$$Re(U) = \begin{pmatrix} -0.0478 & 0.4977 & 0.0000 & 0.0000 \\ -0.2874 & -0.0276 & -0.3055 & -0.7572 \\ -0.2874 & -0.0276 & 0.8085 & 0.1140 \\ -0.2874 & -0.0276 & -0.5030 & 0.6432 \end{pmatrix}, \text{ four-mode star-type} \quad (S14)$$

$$Re(U) = \begin{pmatrix} 0.1590 & -0.0059 & 0.0549 & 0.3657 & 0.1651 & -0.6528 \\ -0.0107 & -0.2866 & 0.4560 & -0.0684 & 0.2905 & 0.0735 \\ 0.3574 & -0.0134 & 0.0305 & 0.2029 & -0.1324 & 0.5235 \\ -0.0134 & -0.3574 & -0.2029 & 0.0305 & -0.5235 & -0.1324 \\ 0.2866 & -0.0107 & -0.0684 & -0.4560 & 0.0735 & -0.2905 \\ -0.0059 & -0.1590 & -0.3657 & 0.0549 & 0.6528 & 0.1651 \end{pmatrix}, \text{ six-mode linear-type} \quad (S15)$$

This theoretical prediction has been supported by experimentally measured correlation matrix of nullifiers in Fig.5c, showing nearly identical diagonal correlations and highly suppressed off-diagonal correlations. The theoretically predicted and experimentally measured squeezing strengths for generating various multipartite entangled states are provided in Table S1. The results suggest that the experimental squeezing strength is nearly proportional to the theoretical values.

entanglement structure	theoretical squeezing strength (D)	experimental squeezing strength ($\text{diag}(e^{-R_l})$)	$1 - \text{cosine-similarity}$	λ
	{0.276, 0.276, 0.724, 0.724}	{0.179, 0.179, 0.462, 0.462}	$1.373 * 10^{-5}$	0.643
	{0.200, 0.200, 1.000, 1.000}	{0.127, 0.128, 0.618, 0.616}	$1.853 * 10^{-5}$	0.626
	{0.250, 0.250, 1.000, 1.000}	{0.141, 0.140, 0.560, 0.560}	$1.373 * 10^{-6}$	0.561
	{0.235, 0.235, 0.391, 0.391, 0.835, 0.835}	{0.143, 0.142, 0.232, 0.232, 0.494, 0.494}	$1.301 * 10^{-5}$	0.596

Table S1 Theoretical and experimental squeezing strength for the preparation of multipartite entanglement with cluster-type structures. The theoretical squeezing strength is calculated by Eq.S9 and the experimental squeezing strength is measured for the selected eigen-supermodes, to generate corresponding multipartite entanglement with cluster-type structures. We use the cosine-similarity (defined as $\mathbf{v}_{thy} \cdot \mathbf{v}_{exp} / (||\mathbf{v}_{thy}|| * ||\mathbf{v}_{exp}||)$, where $\mathbf{v}_{thy}$ and $\mathbf{v}_{exp}$ are the diagonal terms of two diagonal matrices, D and $\text{diag}(e^{-R_l})$, respectively) to quantify the degree to which the experimental squeezing strength is proportional to the theoretical values. When the cosine similarity equals 1, it indicates that the two squeezing strengths are perfectly proportional to each other. The predicted constant λ is defined as the average value of the ratio of each squeezing strength $\text{diag}(e^{-R_l})/D$. The high agreement between the theoretical predictions and the experimental results suggests that the pump configuration has been adjusted effectively to generate multipartite entangled states with cluster-type structures with minimal off-diagonal nullifier correlations.

2.2 Modeling of quantum frequency-multiplexed Gaussian states in the microcomb

Figure 1 shows the scheme for the generation of continuous-variable multipartite entanglement of quantum frequency supermodes. The scheme involves the generation of a mass of quantum frequency modes in an integrated-optical microresonator-based microcomb. Complex quadrature correlations of these modes are established by the polychromatic bright pump beams with a uniform frequency space that are several times of the microresonator FSR. The interaction between bright pump beams and below-threshold vacuum modes can be described by a simplified Hamiltonian under the mean-field approximation:

$$\begin{aligned}
\hat{H}_{int} &= -\hbar\kappa \left\{ \sum_{i,j,k,l}^{\text{SPS}} \delta_{\text{SPS}} (A_i A_j \hat{a}_k^\dagger \hat{a}_l^\dagger + h.c.) + \sum_{i,j,k,l}^{\text{BS}} \delta_{\text{BS}} (A_i^* A_j \hat{a}_k^\dagger \hat{a}_l + h.c.) \right\} \\
&= \frac{\hbar}{2} \sum_{k,l}^{\text{SPS}} [G_{kl} \hat{a}_k^\dagger \hat{a}_l^\dagger + H.c.] + \hbar \sum_{k,l}^{\text{BS}} T_{kl} \hat{a}_k^\dagger \hat{a}_l.
\end{aligned} \tag{S16}$$

The interaction can be separated into two nonlinear optical processes: spontaneous pair squeezing (SPS) and Bragg scattering (BS), described by the coefficient matrices G and T , respectively, which satisfy the condition $G = G^T$, $T = T^T$. The SPS process generates paired quadrature correlations, while the BS process transfers the quadrature correlations to other modes, forming a complex second-order correlation structure in quantum frequency modes.

The dynamics of light fields of below-threshold microcombs can be described by the quantum Langevin equations:

$$\frac{d\hat{\mathbf{x}}(t)}{dt} = (M - \Gamma - \Gamma')\hat{\mathbf{x}}(t) + \sqrt{2\Gamma}\hat{\mathbf{x}}_{in}(t) + \sqrt{2\Gamma'}\hat{\mathbf{x}}_{loss}(t), \quad M = \begin{pmatrix} \Im\{G + T\} & -\Re\{G - T\} \\ -\Re\{G + T\} & -\Im\{G - T\} \end{pmatrix}, \tag{S17}$$

where $\hat{\mathbf{x}} = (\hat{q}_1, \hat{q}_2, \dots, \hat{q}_n | \hat{p}_1, \hat{p}_2, \dots, \hat{p}_n)^T$ is the vector of quadratures, quadratures are defined as $\hat{q}_k = \hat{a}_k^\dagger + \hat{a}_k$, $\hat{p}_k = i(\hat{a}_k^\dagger - \hat{a}_k)$, $\hat{\mathbf{x}}_{in}$ is the quadrature vector of input fields in the bus waveguide, $\hat{\mathbf{x}}_{loss}$ is the quadrature vector of incoming fields through the lossy propagation in the microring. The matrices $M = i[\hat{H}_{int}, \hat{\mathbf{x}}]/\hbar$ and $\Gamma = \text{diag}(\gamma_1, \gamma_2, \dots, \gamma_n | \gamma_1, \gamma_2, \dots, \gamma_n)$, $\Gamma' = \text{diag}(\gamma'_1, \gamma'_2, \dots, \gamma'_n | \gamma'_1, \gamma'_2, \dots, \gamma'_n)$ denotes the gain rate from nonlinear process and damping rate from coupling and loss process, respectively. In the case of over-coupled microresonator, the cavity damping mainly comes from the coupling process. In other words, the quantum Langevin equation can be approximately written as:

$$\frac{d\hat{\mathbf{x}}(t)}{dt} = (M - \Gamma)\hat{\mathbf{x}}(t) + \sqrt{2\Gamma}\hat{\mathbf{x}}_{in}(t). \tag{S18}$$

2.3 Polychromatic homodyne detections and linear manipulation on qumodes

To characterize the quadrature correlations of quantum frequency modes, we implemented polychromatic homodyne detections to measure the second-order correlation function of multi-qumode Gaussian states. As shown in Fig.S3, polychromatic homodyne detection is actually several homodyne detections multiplexed in the frequency domain, which requires only one homodyne setup but can detect correlations for any supermodes. The polychromatic LO beams with different amplitudes $A_i(t)$ and phases ϕ_i provide a well-defined measurement basis for the characterization of multi-qumode Gaussian states. For the case that the frequency response of detectors is flat, the difference of photon currents from two detectors is proportional to the sum quantum noise of each mode:

$$I(t) = I_1(t) - I_2(t) \propto \sum_i A_i(t) (\cos \phi_i \hat{q}_i(t) + \sin \phi_i \hat{p}_i(t)). \tag{S19}$$

Owing to the presence of excess noises at different sideband frequencies, it is necessary to analyze sideband distributions of quadrature squeezing and multipartite entanglement. The power spectral density (PSD) of current difference can be derived from the Wiener-Khinchin theorem:

$$\Sigma_{\{A,\phi\}}^{(I)}(\omega) = \int d\tau e^{-i\omega\tau} \langle I(t+\tau)I(t) \rangle \propto \int d\tau e^{-i\omega\tau} \mathbf{Q}_{\{A,\phi\}}^T \langle \hat{\mathbf{x}}(t+\tau) \hat{\mathbf{x}}^T(t) \rangle \mathbf{Q}_{\{A,\phi\}} \quad (\text{S20})$$

where $\langle \cdot \rangle$ denotes quantum statistical average for stationary stochastic process, $\mathbf{Q}_{\{A,\phi\}} = (A_1 \cos \phi_1, \dots, A_n \cos \phi_n | A_1 \sin \phi_1, \dots, A_n \sin \phi_n)^T$. It should be noted that, for simplicity, we assume that the amplitude $A_i(t)$ of different LOs obeys a Gaussian stochastic process, whose time average is written as A_i . With the addition of normalization, the vector $\mathbf{Q}_{\{A,\phi\}}$ can be easily found as the column vector of the orthonormal symplectic matrix $Q \in Sp(2n, \mathbb{R}) \cap O(2n)$. It means that the implementation of polychromatic homodyne detection is equivalent to linearly manipulating quantum modes and measuring the variance of supermodes.

The multi-qumode entangled states we measured in our experiment are the states coupled out of the chip and directly measured at the homodyne detectors. According to the quantum Langevin equation in Eq.(S18) and the input-output relation $\hat{\mathbf{x}}_{in}(t) + \hat{\mathbf{x}}_{out}(t) = \sqrt{2\Gamma} \hat{\mathbf{x}}(t)$, the transfer matrix function of the entire dynamical process can be derived as [7]:

$$S(\omega) = \sqrt{2\Gamma} (i\omega \mathbb{I}_{2n \times 2n} - M + \Gamma)^{-1} \sqrt{2\Gamma} - \mathbb{I}_{2n \times 2n}, \quad (\text{S21})$$

where the transfer matrix function satisfies $S(\omega)\Omega S^\dagger(\omega) = \Omega$ with $2n \times 2n$ 'symplectic form' Ω . Then the PSD of the measured supermode can be expressed by:

$$\Sigma_{\{A,\phi\}}^{out}(\omega) = \int d\tau e^{-i\omega\tau} \mathbf{Q}_{\{A,\phi\}}^T \langle \hat{\mathbf{x}}_{out}(t+\tau) \hat{\mathbf{x}}_{out}^T(t) \rangle \mathbf{Q}_{\{A,\phi\}} = \mathbf{Q}_{\{A,\phi\}}^T S(\omega) S^\dagger(\omega) \mathbf{Q}_{\{A,\phi\}}, \quad (\text{S22})$$

where the input light field is vacuum, whose PSD is normalized to $\Sigma^{in}(\omega) = \mathbb{I}_{2n \times 2n}$. Notably, for those quantum light sources based on second-order optical nonlinearity, the BS coefficients T vanishes. If different frequency modes possess similar damping rates ($\gamma_i \equiv \gamma$), the multimode PSD matrix is real-symmetric:

$$\Sigma^{out}(\omega) = S(\omega) S^\dagger(\omega) = 4\gamma[(\gamma^2 + \omega^2)M^{-1} + M - 2\gamma]^{-1} + I \in \mathbb{R}^{2n \times 2n}, \quad (\text{S23})$$

which can be diagonalized by the orthonormal symplectic matrix Q . Thus, the quantum noises of eigen-supermode can be measured by the polychromatic homodyne detections. However, in the case of the third-order optical nonlinearity, the multimode PSD matrix is generally Hermitian and cannot be diagonalized by orthonormal symplectic matrices. The information obtained from homodyne detections represents the real part of a genuine multimode PSD matrix:

$$\Sigma_{\{A,\phi\}}^{out}(\omega) = \mathbf{Q}_{\{A,\phi\}}^T \Re\{S(\omega) S^\dagger(\omega)\} \mathbf{Q}_{\{A,\phi\}}. \quad (\text{S24})$$

This intrinsic imperfection of homodyne detections can lead to the difficulty of reconstructing the full multimode PSD matrix. This issue can be overcome by the resonator detection technique [8]. Fortunately, in our experiment, implementing homodyne detections – at broad sideband frequencies, is still sufficient to measure non-optimal eigen-supermodes, as well as to verify multipartite entanglement. Details are discussed in Sec.2.5.

2.4 The impact of Bragg scattering on the physical mechanics of multipartite entanglement generation in the microcomb

The Bragg scattering term shown in the Eq.S16 induced by the third-order nonlinear-process make the case much different from the second-order spontaneous parametric down-conversion (SPDC) squeezers in those pioneering experiments [1–3, 9, 10]. First, in the absence of BS term, the coefficient matrix T in the Eq.S16 vanishes. If the polychromatic pumps satisfy the phase-matching condition [11], there exists a quadrature reference frame in which $\Re(G) = 0$, which leads to a block-diagonal $\Sigma^{out}(\omega)$. In other words, the correlations between q -quadrature and p -quadrature vanish. However, in our experiment, the multimode covariance matrix is not necessarily block-diagonal and we have to apply the CPPD method to reconstruct the whole covariance matrix to correctly characterise the generated multimode Gaussian state. As mentioned in Section 2.3, the Bragg scattering can additionally lead to complicated sideband frequency distribution of multimode PSD matrix, which would be far from the usual real-symmetric covariance matrix (see Section 2.5 for details).

The other result from the presence of BS term is that the generated multimode Gaussian state cannot be directly transformed to a cluster-type state by Gaussian local unitaries (GLUs), which is very important for the leverage of entanglement resources. According to ref. [12, 13], the so-called $\mathcal{H}$ -graph method can map the Gaussian state generated by second-order nonlinear-process to a certain cluster-type state by local Fourier transformation operations [1, 2, 9], which, however, cannot be applied for our case. To show this, we extract the on-chip covariance matrix Σ_0 by taking off the mixed vacuum by loss from the measured covariance matrix. By defining

$$\sigma_{jk} = \begin{pmatrix} \Sigma_{0,jk}^{qq} & \Sigma_{0,jk}^{qp} \\ \Sigma_{0,jk}^{pq} & \Sigma_{0,jk}^{pp} \end{pmatrix} \quad (\text{S25})$$

where the superscript denotes different quadrant of the covariance matrix, we can apply the theorem in ref. [14], which claims that if a given Gaussian state is GLU-equivalent to a cluster state, then $\det(\sigma_{jk}) \leq 0$ for all $j \neq k$. We calculate all 36 $\det(\sigma_{jk})$ for eight-frequency-mode covariance matrix, among which 20 $\det(\sigma_{jk})$ are greater than zero and the maximal value is up to 47.03, showing strong violation of GLU-equivalent condition. That means we cannot use GLU operations (let alone the local Fourier transformation in $\mathcal{H}$ -graph method) to directly transform the generated Gaussian state to a certain cluster-type state. Therefore, we utilize a building-up method that globally operate eigen-supermodes through the linear operations by tailoring the LOs (see Section 2.1 for details).

2.5 Multimode covariance matrix reconstruction and supermodes measurements

To obtain the squeezing degree of non-optimal eigen-supermodes only by homodyne detections, we reconstruct the multimode covariance matrix (the real part of genuine multimode PSD matrix) $\Sigma = \Re\{S(\omega_0)S^\dagger(\omega_0)\}$ at a certain sideband frequency ω_0 and measure the state in the supermode bases by Williamson decomposition and Bloch-Messiah decomposition. We firstly lock the relative phases of polychromatic LOs to give a well-defined reference frame of phase space, in which the LO with initial phase defines q -quadrature base and the LO with $\pi/2$ additional phase defines p -quadrature base. The diagonal terms of the covariance matrix can be determined by a single LO with 0 and $\pi/2$ additional phase and the single-mode $q - p$ correlation can be measured by a single LO with $\pi/4$ additional phase. Moreover, the quadrature correlation between the two modes can be measured by two phase-locked LOs extracted from polychromatic LOs.

After the reconstruction of the covariance matrix from correlation measurements (as shown in Fig.S6), we apply the Williamson decomposition to obtain the symplectic transformation S and conduct the Bloch-Messiah decomposition for S :

$$S = O_1 K O_2, \quad (\text{S26})$$

where $O_1, O_2 \in Sp(2n, \mathbb{R}) \cap O(2n)$ are orthonormal symplectic matrices, and $K = \text{diag}(k_1, \dots, k_n |k_1^{-1}, \dots, k_n^{-1})$ denotes the squeezing degree. The supermode bases are just the column vectors of the orthonormal symplectic matrix O_1 , which can be obtained by the polychromatic homodyne detections discussed in Sec.2.3.

The standard characterization of noise levels using the homodyne detection is set at a sideband frequency. Previous experiments revealed that the squeezing strength is dependent on the choose of sideband frequency, e.g, in Refs. [15, 16]. For multimode squeezing and entanglement, previous theoretical work showed that in general nonlinear processes in the presence of the Bragg scattering, the covariance matrix at high sideband frequencies can be far from real-symmetric but Hermitian and cannot be fully captured by the homodyne detection [7]. According to this proposal, we compared the measured covariance matrix at different sideband frequencies using the homodyne detection to those based on the theoretical model (shown in Fig.3b), and we estimated the truly squeezing degree of supermodes derived by the complex Hermitian covariance matrix (shown in Fig.S8). To the best of our knowledge, we for the first time confirm that the theory in Ref. [7] can precisely model the 3rd-order FWM squeezers, and we also experimentally confirmed that low sideband frequencies (e.g, down to 5MHz in our experiment) was necessary to correctly detect the squeezing degree of eigen-supermodes. In Fig.S8, we compare the squeezing degree by using optimal and non-optimal eigen-supermodes. It shows that optimal supermode squeezing obtained from the PSD matrix and non-optimal supermode squeezing fitted by experimental data, is nearly identical at the 5 MHz basic sideband frequency (we adopted in our experiment), and they are still close to each other at the whole sideband frequencies ranging from 5 MHz to 500 MHz. In other words, in the third-order microcomb in the presence of Bragg scattering, only if we take the approximately optimal sideband frequency can we acquire the approximately correct results by using the homodyne detections. In contrast, in the 2nd-order SPDC squeezers in the absence of Bragg scattering, the multimode covariance matrix at different sideband frequencies can be approximately real-symmetric (with similar damping rate for all modes, see Section 2.3), so that the matrix can be fully captured by the homodyne detection. These results thus validate the sufficiency of implementing homodyne detections for the experimental characterizations of quadrature squeezing and multipartite entanglement at those sideband frequencies.

3 Characterization of multi-qumode entanglement generated in the microcombs

3.1 Positive partial transposition criteria for continuous-variable quantum systems

The positive partial transposition (PPT) criteria are often used to determine whether two subsystems of a composite system are separable or not. For example, if subsystem A and B are separable, i.e, $\rho_{AB} = \rho_A \otimes \rho_B$, the partial transposition of subsystem A, $\rho_{AB}^{T_A} = \rho_A^T \otimes \rho_B$ (or equivalently the partial transposition of subsystem B, $\rho_{AB}^{T_B} = \rho_A \otimes \rho_B^T$) is also a physical state. We can obtain a sufficient inseparability criterion for bipartite entangled systems, by checking the violation of physical-state requirement of the partially transposed state.

For a CV Gaussian state, the PPT criteria can be fully characterized by its covariance matrix Σ . In this case, the transposition of density operator corresponds to the negation of all momentum operators in the phase space:

$$\mathbf{x} \rightarrow \mathbf{x}' = (q_1, q_2, \dots, q_n, -p_1, -p_2, \dots, -p_n). \quad (\text{S27})$$

The effect of the partial transposition on subsystem A when applied to the covariance matrix is to negate the matrix elements that contain the momentum operator p_j ($j \in \text{subsystem A}$) exactly once. We denote the partially transposed covariance matrix as $\tilde{\Sigma}$. For the covariance matrix, there is also a requirement for physical state (equivalent to the requirement of $\rho \geq 0$ for density operator):

$$\Sigma + i\Omega \geq 0, \quad (\text{S28})$$

where $\Omega = \begin{pmatrix} 0 & \mathbb{1}_{N \times N} \\ -\mathbb{1}_{N \times N} & 0 \end{pmatrix}$ is $2n \times 2n$ symplectic form. If subsystem A and B are separable, then $\tilde{\Sigma}$ is also a physical covariance matrix satisfying:

$$\tilde{\Sigma} + i\Omega \geq 0. \quad (\text{S29})$$

This condition is equivalent to the following: covariance matrix with symplectic eigenvalues no less than 1. In other words, we can conduct the Williamson decomposition to the partial transposed covariance matrix $\tilde{\Sigma}$,

$$\tilde{\Sigma} = S^T \nu S, \quad \nu = \text{diag}(v_1, v_1, v_2, v_2, \dots, v_n, v_n), \quad (\text{S30})$$

and the separable condition corresponds to all $v_i \geq 1$ for $i = 1, 2, \dots, n$. Therefore, if $\tilde{\Sigma}$ contains symplectic eigenvalue smaller than 1, then the subsystems A and B are inseparable.

For the eight-qumode Gaussian states, generated in the integrated microcomb, there are a total of 127 ways to divide the eight-qumode system into two subsystems, as shown in Fig.S7a, including bipartition of 1×7 (bipartition 0 ~ 7), 2×6 (bipartition 8 ~ 35), 3×5 (bipartition 36 ~ 91), and 4×4 (bipartition 92 ~ 126). We calculate the complete symplectic spectra for all these 127 ways of partial transposition and take out the minimum symplectic eigenvalue for each symplectic spectrum, as shown in Fig.S7a. Among these 127 bipartitions, the smallest symplectic eigenvalue appears in the bipartition 112 (off-chip $\nu_{\min} = 0.577$ and on-chip $\nu_{\min} = 0.067$ at 5 MHz sideband frequency), where subsystems A contains the frequency modes of $\{-5, -2, 1, 4\}$ and B contains the frequency modes of $\{-4, -1, 2, 5\}$. The on-chip symplectic eigenvalues are estimated by inclusion of the detection loss of $\eta = 0.473$. The result shows the presence of strong entanglement between these two subsystems. In Fig.S7b, we characterize symplectic eigenvalues of the 127 bipartitions at different sideband frequencies. Although symplectic eigenvalues slightly increase at the higher sideband frequencies, the separable criterion for symplectic eigenvalues larger than one is always violated. This result demonstrates the presence of multipartite entanglement at broad sideband frequencies.

3.2 Violations of van Loock-Furusawa inequalities

The nullifiers of a continuous-variable cluster state are defined as:

$$(\hat{p}_i - \sum_j \mathcal{V}_{ij} \hat{q}_j) \rightarrow 0, \quad (\text{S31})$$

where $i, j \in G$, G is a graph in which vertexes represent qumodes, and the adjacent matrix $\mathcal{V}$ of the graph determines quadrature correlations. One can prepare multipartite entangled states with different cluster-type structures by using single-mode squeezing and controlled two-mode operations, which can be decomposed to several single-mode squeezers operated by linear-optic transformation [6]. In our experiment, the eigen-supermodes serve as single-mode squeezers and the linear-optic transformation is equivalently realized by performing the polychromatic homodyne detections [3]. By tailoring the amplitudes and phases of polychromatic LO beams, we reconfigure entangled states with different entanglement structures, as shown in Fig.4b. To characterise the entanglement structure of generated states, we can measure the nullifiers for every vertices in the cluster: $\mathcal{N}_i = \hat{p}_i - \sum_j \mathcal{V}_{ij} \hat{q}_j$. We find that for any edge in the cluster, the sum of involved nullifiers all violate the van Loock-Furusawa inequalities:

$$\mathcal{LF} = \langle (\Delta \mathcal{N}_i)^2 \rangle + \langle (\Delta \mathcal{N}_j)^2 \rangle \geq 4. \quad (\text{S32})$$

which suggests the existence of quantum entanglement for all generated states. The results are shown in Fig.4c. A full list of inequalities for different multipartite states and the experimentally measured $\mathcal{N}_i$ and $\mathcal{LF}$ values have been provided in Table S2.

3.3 Characterizations of multipartite entanglement with cluster-type structures by measuring the nullifier correlations

As discussed in Section 2.1 and Section 2.4, due to the presence of BS effect in third-order nonlinear-process, we utilize a improved building-up method based on the scheme in ref.[6] to generate multipartite entanglement with cluster-type structures, by experimentally tailoring the configurations of polychromatic pump beams and polychromatic LO beams.

Firstly, using only linear unitary operations could lead to unexpected off-diagonal nullifier correlations in the finite squeezing region [6]. The crux of the issue lies in the fact that this method only ensures the elimination of divergent noise terms caused by the antisqueezing of squeezers, resulting in the condition in Eq.S6. Under this condition, the correlation matrix of nullifiers defined by A can be written as Eq.S8, which is not necessarily diagonal for finite squeezing e^{-R_l} . For instance, when all squeezers have same squeezing strength (e.g, $R_l \equiv R$), which is common in CV-related experiments, from Eq.S7 we can obtain:

$$\begin{aligned} \text{cov}[\hat{p}_c - A \cdot \hat{q}_c] &= e^{-R} * (Re(U) + A \cdot Im(U)) \cdot (Re(U) + A \cdot Im(U))^T \\ &= e^{-R} * (I + A^2) Re(U) Re(U)^T (I + A^2) \\ &= e^{-R} * (I + A^2), \end{aligned} \quad (\text{S33})$$

which exhibits intrinsic off-diagonal terms induced by A^2 that are in the same order of the diagonal terms unless A is self-inverse. Although the left hand side of Eq.S8 asymptotically vanishes when the squeezing strength goes to infinity, the unexpected off-diagonal nullifier correlations indicates the existence of hidden entanglement not accounted for by the anticipated cluster structure A [14]. Therefore, to prepare entanglement structure that closely approximates the CV cluster structure, additional conditions on the linear operation U and the squeezing strength e^{-R_l} are necessary to eliminate the off-diagonal terms in Eq.S8.

Combining the condition in Eq.S6, and our orthogonality condition in Eq.S9, allows to uniquely determine the U and give the required theoretical squeezing strength D . This additional condition in Eq.S9 could force the correlation matrix of nullifiers into an identity matrix multiplied by a certain constant λ , which leads to a good approximation of multipartite entanglement with cluster-structures. We arrive at this condition by finely tailoring the intensity and detuning of the polychromatic pump for each cluster-type structure in our experiment, resulting in highly suppressed off-diagonal nullifier correlations (shown in Fig.5) and near identical diagonal terms close to the theoretical value of λ (shown in Table S1). In experiment, in order to approach the theoretical squeezing strength, we engage in a trade-off between the total squeezing strength and the distribution of squeezing degrees among the squeezers. For the same reason, we choose different squeezers (eigen-supermodes) for each cluster-type structures, such as $\{Q_{e3}, Q_{e4}, Q_{e5}, Q_{e6}\}$ for the four-mode linear-type entangled state, $\{Q_{e1}, Q_{e2}, Q_{e5}, Q_{e6}\}$ for the four-mode star-type and box-type entangled states, and $\{Q_{e1}, Q_{e2}, Q_{e3}, Q_{e4}, Q_{e5}, Q_{e6}\}$ for the six-mode linear-type entangled state.

Moreover, since the presence of losses can introduce off-diagonal correlations by mixing the vacuum into the on-chip generated state:

$$\text{cov}[\hat{p}_c - A \cdot \hat{q}_c]_{\text{off-chip}} = \eta * \text{cov}[\hat{p}_c - A \cdot \hat{q}_c]_{\text{on-chip}} + (1 - \eta) * (I + A^2), \quad (\text{S34})$$

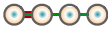
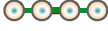
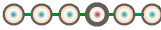
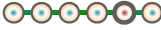
cluster supermode (Q_c)	variance of nullifiers ($\mathcal{N}_i$)	entanglement edge	van Loock-Furusawa values
	$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2})^2 \rangle = 1.349(7)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2})^2 \rangle + \langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 3.223(13)$
	$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 1.874(10)$		$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle + \langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 3.748(17)$
	$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 1.874(10)$		$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,3})^2 \rangle = 3.221(13)$
	$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,3})^2 \rangle = 1.347(8)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 3.727(14)$
	$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 1.869(10)$		$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle + \langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 3.721(11)$
	$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 1.858(10)$		$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 3.723(13)$
	$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 1.863(8)$		$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle + \langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 3.729(14)$
	$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 1.860(9)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,3} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1})^2 \rangle = 3.661(13)$
	$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,3} - \hat{q}_{c,4})^2 \rangle = 2.353(11)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,3} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,1})^2 \rangle = 3.665(12)$
	$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1})^2 \rangle = 1.308(7)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2} - \hat{q}_{c,3} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,1})^2 \rangle = 3.668(11)$
	$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,1})^2 \rangle = 1.312(6)$		$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2})^2 \rangle + \langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 3.178(10)$
	$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,1})^2 \rangle = 1.315(7)$		$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle + \langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 3.711(15)$
	$\langle \Delta(\hat{p}_{c,1} - \hat{q}_{c,2})^2 \rangle = 1.325(7)$		$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle + \langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,3} - \hat{q}_{c,5})^2 \rangle = 3.709(13)$
	$\langle \Delta(\hat{p}_{c,2} - \hat{q}_{c,1} - \hat{q}_{c,3})^2 \rangle = 1.853(9)$		$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,3} - \hat{q}_{c,5})^2 \rangle + \langle \Delta(\hat{p}_{c,5} - \hat{q}_{c,4} - \hat{q}_{c,6})^2 \rangle = 3.705(13)$
	$\langle \Delta(\hat{p}_{c,3} - \hat{q}_{c,2} - \hat{q}_{c,4})^2 \rangle = 1.858(12)$		$\langle \Delta(\hat{p}_{c,5} - \hat{q}_{c,4} - \hat{q}_{c,6})^2 \rangle + \langle \Delta(\hat{p}_{c,6} - \hat{q}_{c,5})^2 \rangle = 3.177(12)$
	$\langle \Delta(\hat{p}_{c,4} - \hat{q}_{c,3} - \hat{q}_{c,5})^2 \rangle = 1.851(7)$		
	$\langle \Delta(\hat{p}_{c,5} - \hat{q}_{c,4} - \hat{q}_{c,6})^2 \rangle = 1.854(11)$		
	$\langle \Delta(\hat{p}_{c,6} - \hat{q}_{c,5})^2 \rangle = 1.323(6)$		

Table S2 Experimental measurement of nullifiers and the van Loock-Furusawa values for the multipartite entangled states with different structures. The nullifiers were chosen according to the graph structures of the cluster-type states, and they were directly measured in the experiment by polychromatic homodyne detections. The values of van Loock-Furusawa inequalities for the edges in approximate cluster-type states were obtained by summing the variances of corresponding nullifiers. If the measured value $\mathcal{LF}$ is less than 4, the inequality is violated, and if the inequalities for all edges are violated, it suggests the presence of multipartite entanglement in the generated states. For the four-qumode linear-type state, all three inequalities have been successfully violated; for the four-qumode box-type state, all four inequalities have been successfully violated; for the four-qumode star-type state, all three inequalities have been successfully violated; for the six-qumode linear-type state, all five inequalities have been successfully violated. These results thus demonstrates the presence of quantum entanglement in all generated multimode states.

we utilized the on-chip covariance matrix of nullifiers to accurately characterize the generated states, effectively isolating the impact of coupling losses from our analysis. The measured nullifier correlations could still deviate from the theoretical value λI , which mainly stems from the errors in setting the intensity of LO beams through wave-shaper. Hence, through the measurement of correlation matrices of nullifiers with low-noise off-diagonal correlations, combined with the experimental violations of van Loock-Furusawa inequalities, we characterised the entanglement structure for approximate cluster-type states.

4 Discussions on scalability, squeezing, fabrication and hybridisation of CV integrated quantum photonics

The number of entangled qumodes created on a photonic chip is currently limited and the level of squeezing measured in this study is rela-

tively modest, compared to advanced bulk-optic and fiber-optic systems. However, for the first integrated-optic CV multipartite entangled states, the crucial focus lies in fully grasping the underlying physics and rigorously validating their authenticity as multipartite entangled states. Building upon our initial demonstration in the current work, both the number of entangled qumodes and the degree of squeezing can be enhanced on the integrated photonics platform.

Scalability: The technology is now poised for the on-chip generation of hundreds or even thousands of entangled qumodes in a single microcomb, achieved by dispersion engineering. For instance, a microcomb with an extended circumference of 0.895m [17], or the utilization of octave-spanning microcombs [18, 19], both have been demonstrated in the SiN platform, can generate approximately 10,000 frequency modes. This is complemented by advanced polychromatic pumping and detection technologies, that feature remarkable scalability. Alternatively, harnessing the bright combs within solution crystals may offer an efficient method for generating cluster-type states [20]. Moreover, we visualize the potential to escalate this capability to cluster-type states comprising millions (even billions) of qumodes by integrating a multitude of path and frequency (even time) multiplexed qumodes across numerous microcombs and circuits. Realizing this vision will certainly necessitate the development of more sophisticated pumping and detection technologies.

Squeezing: The level of squeezing is highly reliant on the design and fabrication of the microcomb. For instance, optimizing the coupling between the microresonator and waveguide so as to optimize the escaping efficient of pump and generated squeezed beams, have a significant impact on the squeezing performance. Our recent findings show approximately 3dB of measured squeezing and approximately 10dB of on-chip squeezing in a separate study. By enhancing the linear loss of SiN waveguides and improve the Q-factor [17], the intrinsic on-chip squeezing capability can be further elevated by up to 15dB in SiN micro-resonators [21].

Fabrication: The Si-based platform play a pivotal role in achieving integrated-optic CV devices with scalability and stability. The remarkable capabilities of very-large-scale integration of photonic circuits on a Si/SiN platform, along with the integration of multimode squeezing microcombs, linear-optic quantum circuits, and balanced homodyne detectors, offer a highly promising avenue for enabling the generation and processing of large-scale qumode cluster-type states. Recent advancements in fabrication technologies have demonstrated the feasibility of integrating squeezers, circuits, and photodiodes into a single chip system [22, 23]. This integrated approach holds the potential to significantly enhance the strength of measurable squeezing. Furthermore, it opens up possibilities for the precise preparation, control, and detection of individual qumodes, as well as the deterministic generation and precise manipulation of entangled qumodes, all within a unified chip architecture.

Hybridization: The field of DV integrated quantum photonics has seen significant advancements, encompassing high-quality photon-pair sources, programmable circuits, and single-photon detectors [24–26]. Our current work addresses a critical gap in CV integrated quantum photonics technologies. Crucially, as we strive towards the realization of universal continuous-variable quantum computing and various other quantum photonic applications, the integration of hybrid quantum photonic technologies combining DV-CV approaches becomes imperative. Techniques such as non-Gaussian operations and the preparation of GKP qubits [27–29], underscore the necessity of such hybrid approaches. Our demonstration of CV multipartite entangled states here not only stands on par with our previous achievements in DV multiphoton chips on the Si-based platform but also paves the way for the realization of DV-CV hybrid quantum photonics on integrated photonic chips.

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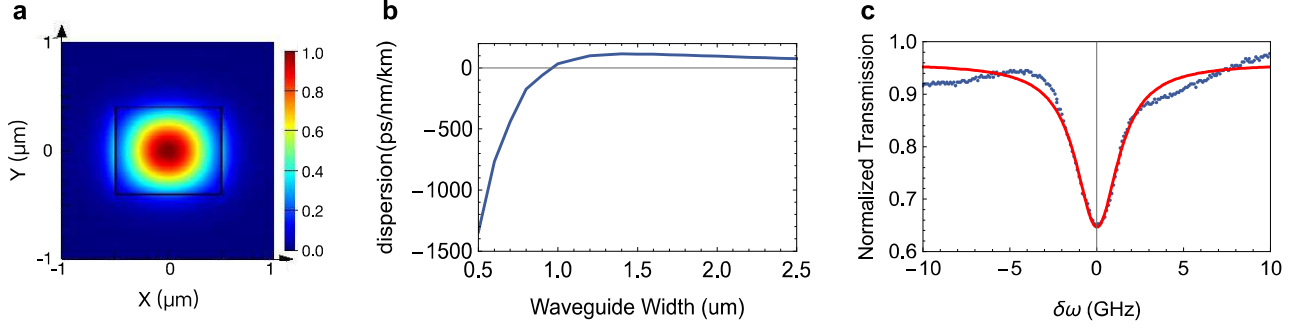


Figure S1 Design of silicon nitride waveguide and microresonator. (a), Simulated field distribution for the transverse electric (TE) fundamental mode in a single-mode silicon nitride waveguide with a width of 1000 nm and thickness of 800 nm. The Finite Difference Method (FDM) was used for numerical mode simulation. (b), Numerical simulation of structural dispersion for different waveguide widths. The thickness is chosen as 800 nm. We chose the waveguide width of 1000 nm, that provides a small anomalous dispersion for efficient microcomb generation. (c), Experimental calibration of microring quality factors. The intrinsic quality factor was measured to be around 1 million, and the escape efficiency was measured to be about 0.9.

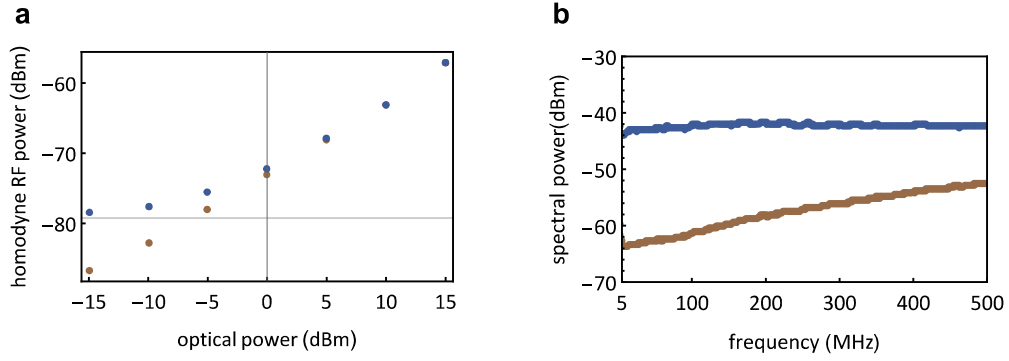


Figure S2 Characterizations of local oscillator noises. (a), Shot noise calibration of local oscillators. The intensity noises of LO were characterized directly by splitting the LO at a 50/50 beamsplitter and detecting the outgoing beams with a balanced photodetector. We characterized the total noises of the LO by measuring the intensity noises at each photodiode separately. By subtracting the electronic noises of the spectrometer and detector, we obtained a nearly linear relationship between the shot noises and the LO optical power. We selected the LO power to be 10 dBm in all measurements, so that the proportion of the electronic noises in the shot noises was small enough, and in the linear region, to avoid measurement inaccuracies caused by detector saturation. (b), Calibration of the common mode rejection ratio. In the frequency range of 5 MHz–500 MHz, the common mode rejection ratio of > 10dB was measured. In our system, although the common-mode signal has been subtracted, there was still a lot of noise at the frequencies lower than 3 MHz. We choose 5 MHz as the basic sideband frequency to characterize squeezing and entanglement.

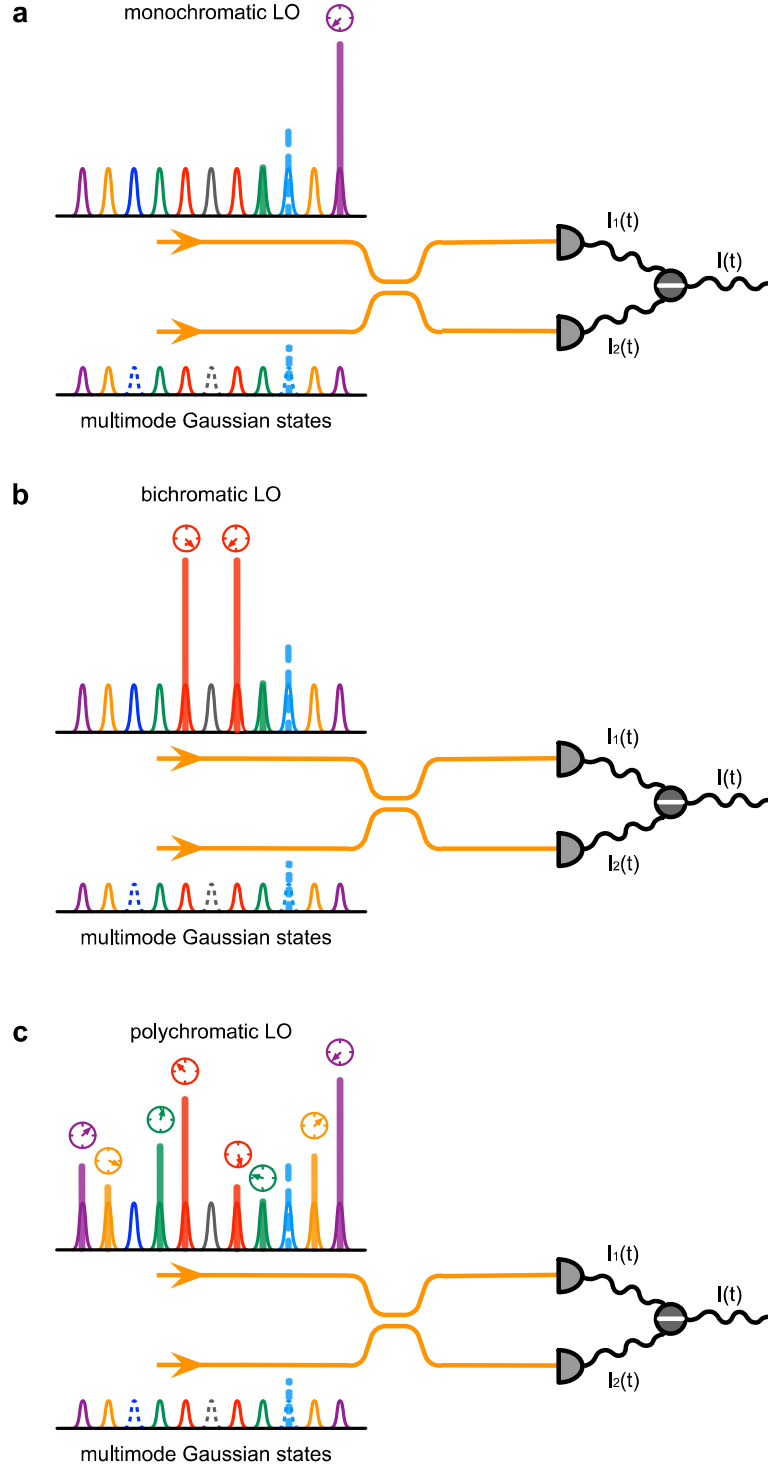


Figure S3 Schematic diagrams for different homodyne detections. (a), Monochromatic, (b), bichromatic, and (c), polychromatic homodyne detections. Local oscillators (LOs) with different amplitudes (indicated by heights of combs) and relatively stable phases (indicated by pointers above combs) in each frequency comb-mode can be arbitrarily reconfigured using the wave-shaper. The LO beams interfere with the multimode Gaussian beams on a balanced beamsplitter, and the second-order correlations between different modes can be extracted from the photon current difference $I(t) = I_1(t) - I_2(t)$. Monochromatic homodyne detections were implemented to calibrate the short noise levels in each mode; bichromatic and monochromatic homodyne detections were implemented to characterize the covariance matrices in the quantum frequency comb-modes; polychromatic homodyne detections were implemented to characterize and verify the multipartite entanglement of supermodes that can be regarded as the superposition of frequency comb-modes. To fully measure the covariance matrix of the generated Gaussian states in the microcomb, we used the CPPD method that employs the f_{+3} (indicated by the blue dashed line) beam as the reference. In the CPPD, as the multi-pump beams propagate with multimode squeezed beams along the same optical fiber, from the excitation, to on-chip microcomb, and to detection (as depicted by the bottom path in (c)), the relative phases between the pump and squeezed beams are stable. Similarly, the polychromatic LO beams and the reference beam (f_{+3} beam) also co-propagate along the same fiber (as depicted by the top path in (c)), enabling the preparation of reliable LOs and stabilizing the relative phase between LO and f_{+3} beams. The reference f_{+3} beam, extracted after the beam splitter (the beam splitter in the homodyne detection closes the interferometer), is recycled to achieve a stable overall PID-locked phase between LOs and multimode squeezed beams.

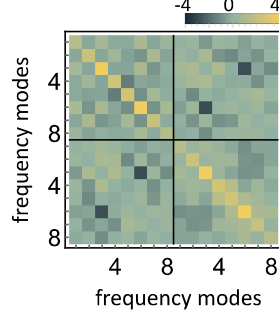


Figure S4 Measured covariance matrix for the eight frequency modes. Measured covariance matrix of the original frequency modes reveals complicated correlations, particularly the presence of $q - q$ off-diagonal correlations, which stem from the intrinsic BS process in the Kerr microcomb, as detailed in Supplementary Section 2.4. The presence of off-diagonal correlations implies that cluster-type states cannot be directly generated in the frequency modes through any Gaussian local unitaries. Instead, the generation of cluster-type states occurs within the eigen-supermodes of the microcomb, enabled by non-local multimode operations. Experiment data was collected at the 5 MHz sideband frequency.

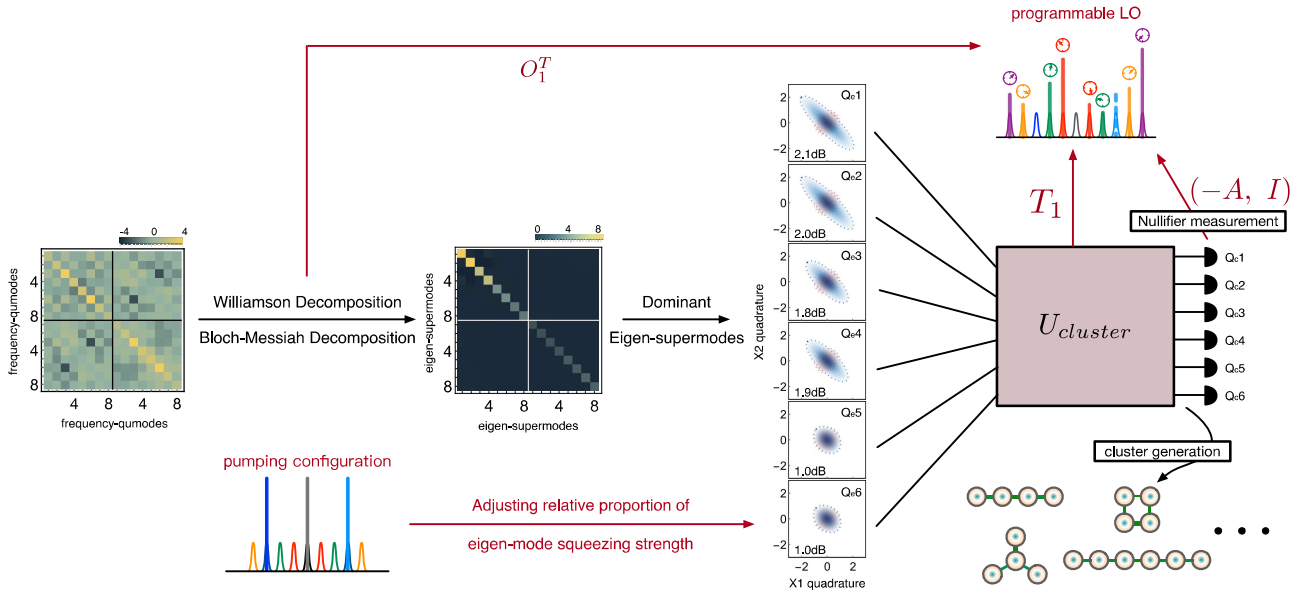


Figure S5 A flow diagram for the generation, manipulation, measurement and characterization of supermode multipartite entanglement with various cluster-type structures. A step-by-step flow diagram for the generation of supermode multipartite entangled states with various cluster-type structures in the microcomb, as well as the manipulation, measurement and characterization of those states.



Figure S6 A full set of experimentally measured paired quadrature correlations. Bichromatic homodyne detections were implemented to characterize quadrature correlations between paired frequency combs. All traces have been normalized to the vacuum noise levels that were measured by performing monochromatic homodyne detections. The quadrature components x and p were measured by adjusting the wave-shaper to rotate the LO light with a 0 and $\pi/2$ phase, respectively. Switching of LOs was enabled by operating the wave-shaper, and LOs were adjusted to the same power by controlling the wave-shaper. All data were collected at the 5 MHz sideband frequency.

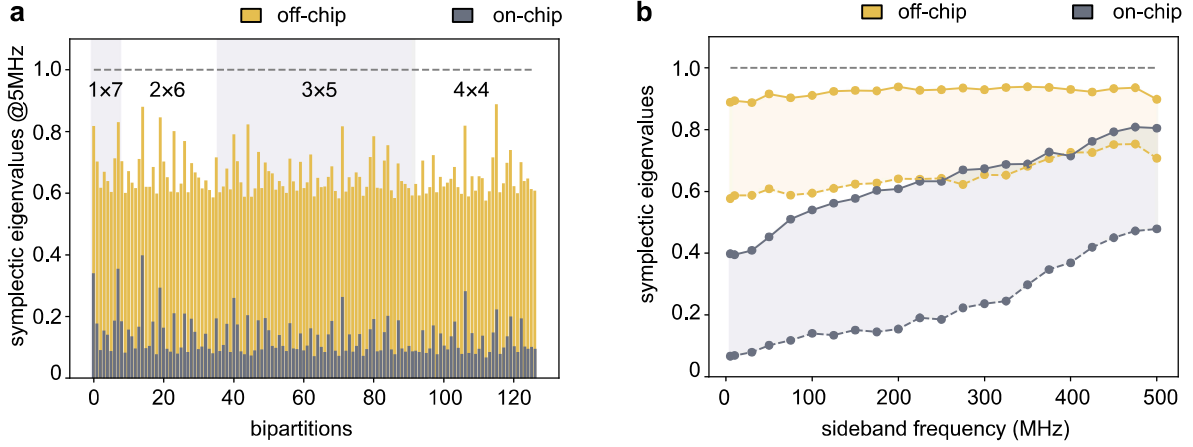


Figure S7 A full characterization of the PPT criteria for eight-qumode entanglement. (a), Experimental violations of the PPT criteria for directly measured eight-qumode Gaussian state (off-chip) and for inferred eight-qumode Gaussian state (on-chip). The symplectic eigenvalues for all 127 bipartitions are below one, in both off-chip and on-chip cases. In (a), experimental data were measured at the 5 MHz basic sideband frequency. (b), Experimental violations of the PPT criteria for the off-chip (yellow) and on-chip (gray) eight-qumode Gaussian state at different sideband frequencies. Symplectic eigenvalues for all 127 bipartitions are below one for both off-chip and on-chip cases, at broad sideband frequencies from 5 MHz to 500 MHz. In (b), the yellow solid (dashed) line refers to the maximum (minimum) symplectic eigenvalues for the off-chip states; the grey solid (dashed) line refers to the maximum (minimum) symplectic eigenvalues for the on-chip states.

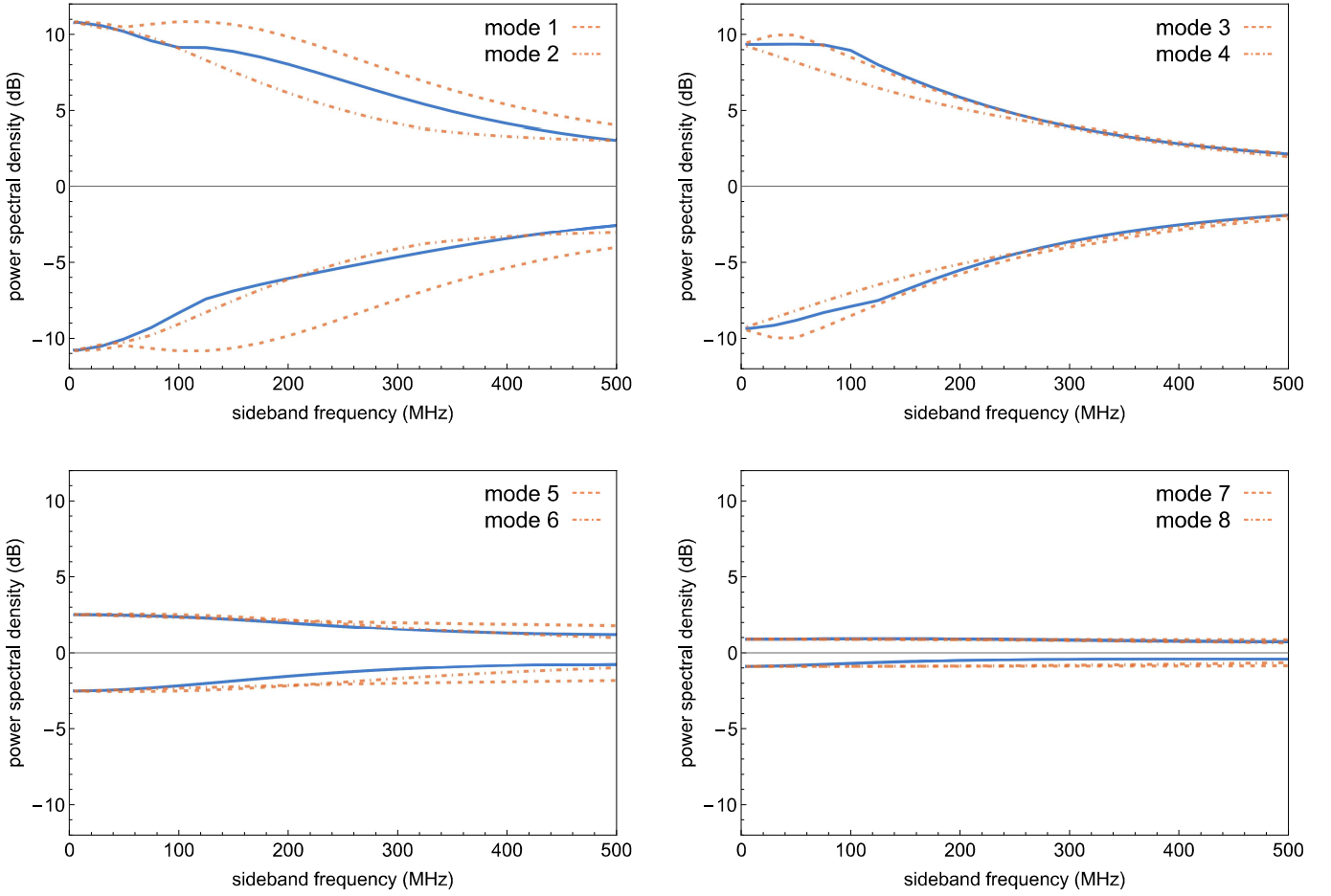


Figure S8 Theoretical estimations of optimal and non-optimal supermode squeezing at different sideband frequencies. The blue solid lines represent the non-optimal supermodes squeezing measured by the homodyne detections (with exclusion of loss after the microresonator), which show the double degeneracy observed in our experiments. Orange dashed and dot-dashed lines represent the optimal supermodes squeezing for the genuine PSD matrix. Blue and orange lines are nearly identical at the 5 MHz basic sideband frequency, and close to each other at the whole sideband frequencies ranging from 5 MHz to 500 MHz. The results shown in the main text Fig.2b refer to directly measured (off-chip) supermodes squeezing at different sideband frequencies.

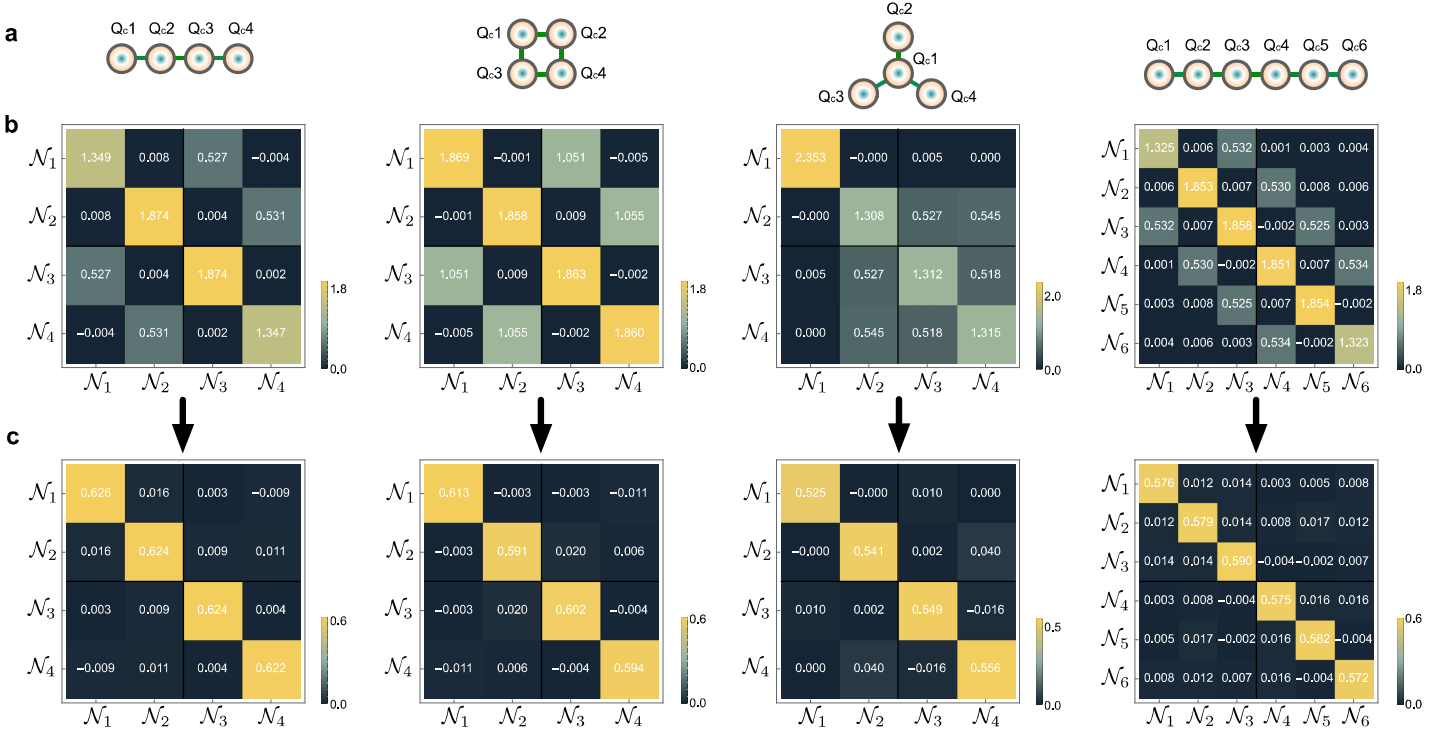


Figure S9 Full characterizations of nullifier correlations. (a), Graph representations for various multipartite entangled states with cluster-type structures. (b), Experimental results of off-chip correlation matrices of nullifiers for corresponding cluster-type structures. Owing to the mixed vacuum, the off-chip states possess off-diagonal nullifier correlations induced by $\text{cov}[\hat{p}_c - A \cdot \hat{q}_c]_{\text{off-chip}} = \eta * \text{cov}[\hat{p}_c - A \cdot \hat{q}_c]_{\text{on-chip}} + (1 - \eta) * (I + A^2)$. (c), Inferred on-chip correlation matrices of nullifiers for corresponding cluster-type structures, when considering optical losses in the measurement system. The diagonally dominant nullifier correlation matrices imply the multipartite entanglement structures are very close to the approximate cluster-type structures in the finite squeezing region. Those results in (c) have been shown in Fig.5 in main text. The elements of correlation matrices is coded by colors according to its absolute value ($|\text{cov}(\mathcal{N}_i, \mathcal{N}_j)|$) and the key is provided at the right bottom. The values of each elements are displayed for clarity.

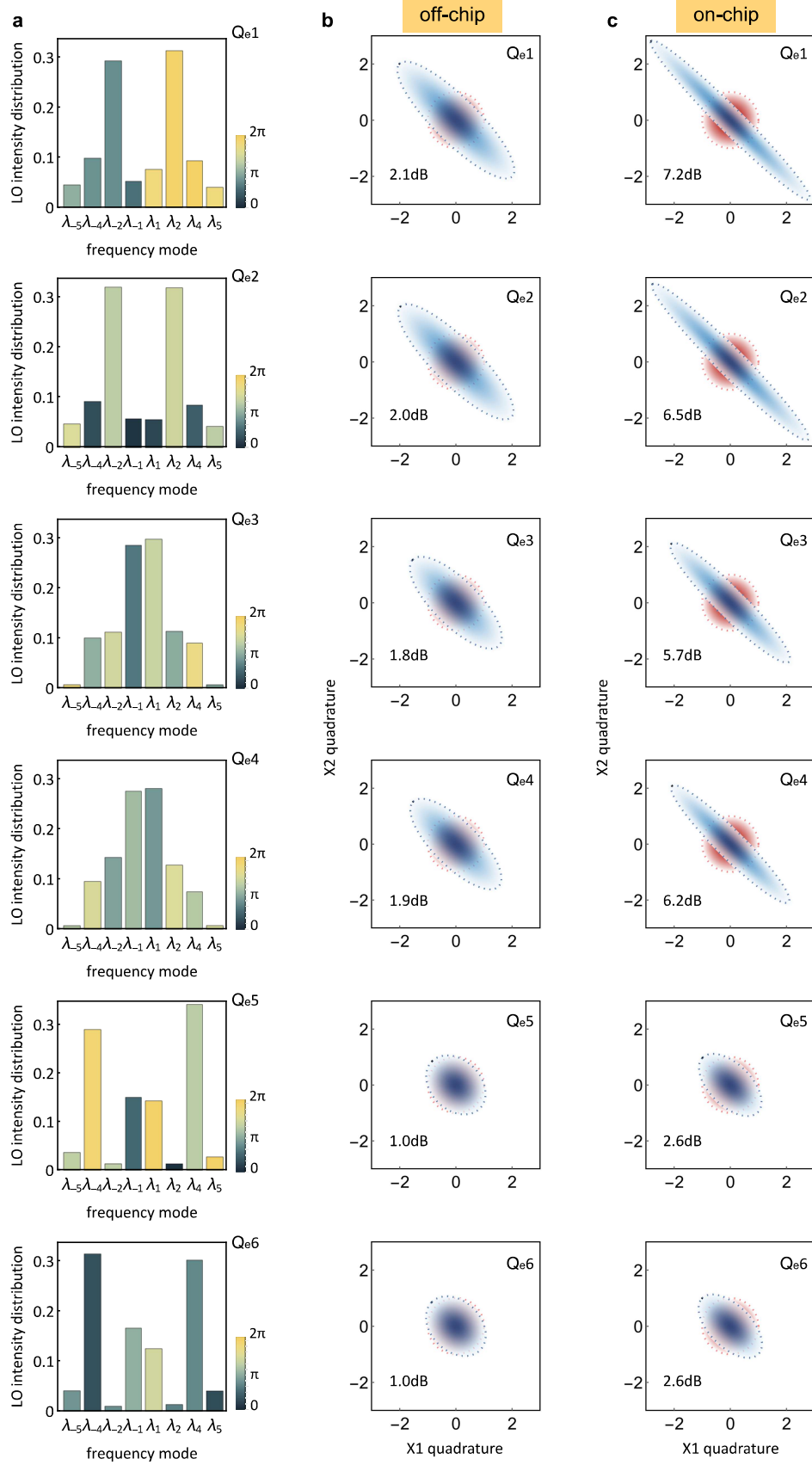


Figure S10 Configurations of polychromatic LO beams for supermode squeezing measurements and estimated phase diagrams for the six supermodes. (a), Intensities and phases of LO beams for the characterizations of quadrature squeezing and anti-squeezing of the six dominated supermodes. Column heights represent the intensities of LOs, and colors represent the phases of LOs (the key is provided at the right bottom). (b) and (c), Phaser diagrams for the six dominant supermodes, that are estimated off-chip (b) and on-chip (c). The total loss of $\eta = 0.473$ measured in the experiment is used to estimate the on-chip covariance matrix as well as quadrature distributions in (c). Blue ellipses represent the quadrature squeezing and anti-squeezing, while red circles represent vacuum fluctuations.

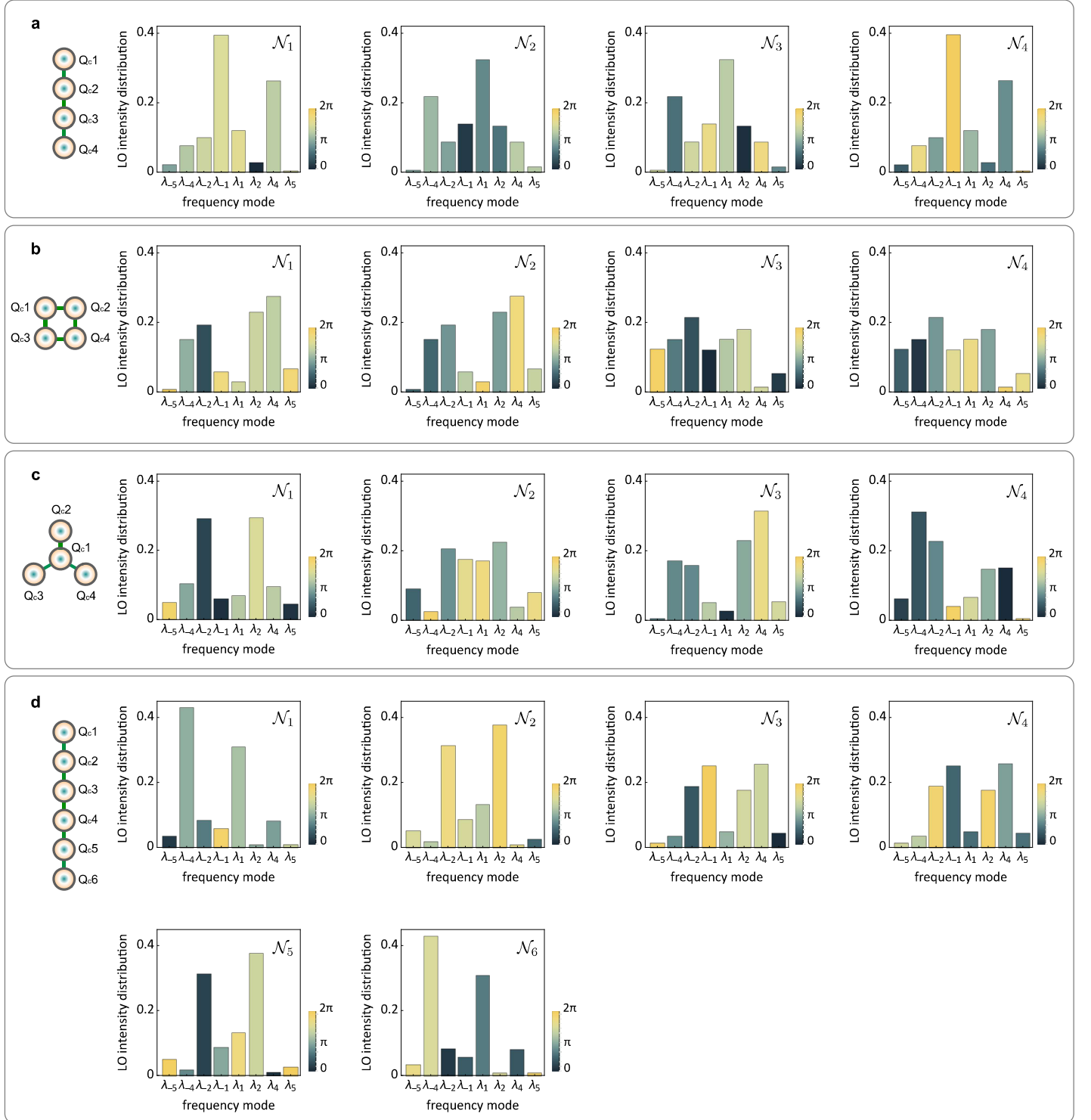


Figure S11 Configurations of polychromatic LO beams for experimental characterizations of nullifiers for different entangled states. Intensities and phases of LO beams for the characterizations of nullifiers for different cluster-type states: (a), four-qumode chain cluster-type states, (b), four-qumode box cluster-type states, (c), four-qumode star cluster-type states, and (d), six-qumode chain cluster-type states. Column heights represent the intensities of LOs, and colors represent the phases of LOs (key is provided at the right bottom).