Gate-tunable frequency combs in graphene-nitride microresonators

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Optical frequency combs, which emit pulses of light at discrete, equally spaced frequencies, are cornerstones of modern-day frequency metrology, precision spectroscopy, astronomical observations, ultrafast optics and quantum information¹⁻⁷. Chipscale frequency combs, based on the Kerr and Raman nonlinearities in monolithic microresonators with ultrahigh quality factors⁸⁻¹⁰, have recently led to progress in optical clockwork and observations of temporal cavity solitons¹¹⁻¹⁴. But the chromatic dispersion within a laser cavity, which determines the comb formation^{15,16}, is usually difficult to tune with an electric field, whether in microcavities or fibre cavities. Such electrically dynamic control could bridge optical frequency combs and optoelectronics, enabling diverse comb outputs in one resonator with fast and convenient tunability. Arising from its exceptional Fermi-Dirac tunability and ultrafast carrier mobility¹⁷⁻¹⁹, graphene has a complex optical dispersion determined by its optical conductivity, which can be tuned through a gate voltage^{20,21}. This has brought about optoelectronic advances such as modulators^{22,23}, photodetectors²⁴ and controllable plasmonics^{25,26}. Here we demonstrate the gated intracavity tunability of graphenebased optical frequency combs, by coupling the gate-tunable optical conductivity to a silicon nitride photonic microresonator, thus modulating its second- and higher-order chromatic dispersions by altering the Fermi level. Preserving cavity quality factors up to 10⁶ in the graphene-based comb, we implement a dual-layer iongel-gated transistor to tune the Fermi level of graphene across the range 0.45–0.65 electronvolts, under single-volt-level control. We use this to produce charge-tunable primary comb lines from 2.3 terahertz to 7.2 terahertz, coherent Kerr frequency combs, controllable Cherenkov radiation and controllable soliton states, all in a single microcavity. We further demonstrate voltage-tunable transitions from periodic soliton crystals to crystals with defects, mapped by our ultrafast second-harmonic optical autocorrelation. This heterogeneous graphene microcavity, which combines singleatomic-layer nanoscience and ultrafast optoelectronics, will help to improve our understanding of dynamical frequency combs and ultrafast optics.

Figure 1a–c shows the concept and fabrication of our graphene gate-tunable Kerr frequency comb with source–drain and top gating. This is further detailed in the Methods and in Supplementary Information sections 1 and 2. To ensure transparency and minimal effect on the resonator quality factor (*Q*) for coherent comb generation, we top-gate the interacting graphene to pull the Fermi level up to 0.6 eV for reduced photon absorption in the nearly massless Dirac cone. An ion-gel capacitor is implemented on top of the graphene monolayer²⁷. The electric double layer in the ionic liquid provides a capacitance up to about $7.2 \,\mu\text{F cm}^{-2}$; this high value enables high doping control

and comb tunability with a few-volt-level gating. This is important to produce sharp modulation of the cavity chromatic dispersion while keeping the cavity loss low. In addition to the optimized 300-nm gap between the Si₃N₄ waveguide and the graphene layer, we optimize the planar interaction length of the arc in which the graphene overlaps the nitride resonator to be about 80 µm. The grey ring shown in Fig. 1a is the nitride resonator. This offers substantial tunability of the frequency comb combined with minimal graphene absorption losses. Figure 1d plots the computed optical group velocity dispersion (β_2) and the computed third-order dispersion (β_3) for tuned Fermi levels from 0.2 eV to 0.8 eV of the graphene monolayer. For each Fermi level, we note the wavelength oscillations in both β_2 and β_3 , arising from the lifetime of the carrier relaxation oscillations in graphene captured in the resonance of the monolayer sheet conductivity. As a result, the graphene β_2 can be tuned from anomalous to normal dispersion and then back to anomalous by means of the gate voltage, which is important for nonlinear phase-matching tunability. This enables wide and tunable frequency comb generation in the graphene-based microresonator (GMR). Based on the modelled overall graphene β_2 and β_3 , we model the heterogeneous microresonator for Kerr frequency comb generation. Figure 1e shows the temporal map of the comb dynamics in the GMR, obtained by Lugiato–Lefever equation (LLE) modelling. At $E_{\rm F} = 0.2 \, {\rm eV}$, the Q factor is low, and hence there is no comb generation. At $E_{\rm F} = 0.5 \, {\rm eV}$, the GMR has $Q \approx 8 \times 10^5$, $\beta_2 \approx -50 \text{ fs}^2 \text{ mm}^{-1}$ and $\beta_3 \approx 0$, resulting in slow comb generation. At $E_{\rm F} = 0.8 \, {\rm eV}$, we observe rapid generation of a full comb in the numerical model, for $Q > 1 \times 10^6$, $\beta_2 \approx -30 \, \text{fs}^2 \, \text{mm}^{-1}$ and $\beta_3 \approx -400 \, \text{fs}^3 \, \text{mm}^{-1}$.

Figure 2a shows the electrical tuning performance of graphene in the GMR. For a fixed source–drain voltage $V_{\rm SD} = 10$ mV, the source– drain current $I_{\rm SD}$ is tuned with the gate voltage $V_{\rm G}$. When $V_{\rm G}$ reaches 2.4 V, $I_{\rm SD}$ has a minimum of 6.5 μ A. Here the carrier density of the graphene monolayer reaches the Dirac point. When $V_{\rm G}$ is less than 2.4 V, graphene is p-doped. In cyclic $V_{\rm G}$ tuning, a hysteresis loop is observed, owing to electronic trapping. The corresponding gate-tunable Fermi energy $|E_{\rm F}| = \hbar |v_{\rm F}| (\pi N)^{-1/2}$ (equation from ref. ²⁸) is plotted in the bottom panel of Fig. 2a and noted to be proportional to $(V_{\rm G})^{1/2}$; here N is the carrier density, while v_F indicates the Fermi velocity. In our experiment, we tune $V_{\rm G}$ in the range -2 V to 0 V, thereby controlling the graphene $|E_{\rm F}|$ between 0.65 eV and 0.45 eV. For $V_{\rm G} = 0$ V, the graphene monolayer in our GMR is already heavily doped, which allows dispersion tuning with low loss.

Figure 2b maps the calculated real and imaginary parts of the GMR, varying with $|E_{\rm F}|$ and wavelength λ . In the two maps, the blue curves denote the boundary where dispersion abruptly changes, and the yellow curve denotes the low-loss region. In our measurement, we apply a high-power continuous-wave pump at 1,600 nm. At this wavelength,

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Fig. 1 | Conceptual design and implementation of the gate-tunable graphene-nitride heterogeneous microcavity. a, Schematic architecture of the GMR, with the silicon nitride indicated in grey. A graphene/ion-gel heterostructure is incorporated in the nitride microresonator. b, Electric-field distribution of the graphene-nitride heterogeneous waveguide, with a Si₃N₄ cross-section of 1.2 × 0.8 μ m². The distance between the Si₃N₄ waveguide and the graphene layer is 100 nm. The graphene and the top-gate probe are separated by 1 μ m with the interlayer ion-gel capacitor. In this structure, transverse electric (TE) mode is applied. c, Optical micrographs show the bus waveguide (red arrows), ring resonator and Au/Ti metallized patterns. An etched window is designed

to ensure both graphene–light interaction and reduced propagation loss. Here the graphene-covered area is marked by the grey dashed box; the etched window label refers to the whole horizontal area between the two central lines. CW, continuous wave; MI: modulated intensity. Scale bar, 100 µm. **d**, Calculated group velocity dispersion and third-order dispersion of graphene, depending on its Fermi level. Here, the curves with $|E_F| = 0.5 \text{ eV}$ and $|E_F| = 0.6 \text{ eV}$, corresponding to the experimental conditions, are highlighted in yellow and red respectively. **e**, Simulated Kerr comb dynamics in the GMR, with different dispersion curves determined by the graphene Fermi level.



Fig. 2 | **Gate-tuning the graphene microring resonator. a**, Electronic measurement of the graphene/ion-gel capacitor. At a source-drain voltage $V_{SD} = 10 \text{ mV}$, the correlation between V_G and I_{SD} shows the Dirac point position and tunable Fermi level of the graphene layer. **b**, Theoretically modelled n_{eff} of the GMR as a function of Fermi levels and optical wavelengths, in which the dispersion and Q can be deduced from the real and imaginary components. Measured data points are shown in white, at a wavelength of 1,600 nm, with $|E_F|$ from 0.5 eV to 0.7 eV.

c, Measured transmissions (top panel) and mode FSR (bottom panel; dots, measured; curves, linear fitting) of the GMR, under gate voltages $V_{\rm G}$ from 0 V to -2 V. **d**, Tuned Q factor and dispersion, under various $V_{\rm G}$. The Q factor increases from 6×10^5 to 1×10^6 as the group velocity dispersion is controlled between -62 fs² mm⁻¹ and +9 fs² mm⁻¹. Error bar is the measurement uncertainty estimated from FSR measurements under the same condition.



Fig. 3 | Observations of the gate-tunable graphene Kerr frequency combs. a, Primary comb lines at controlled gate voltages and Fermi levels of graphene. b, Full frequency combs generated under gate voltages of -1 V, -1.2 V, -1.5 V and -1.8 V. Here the launched pump power is fixed at 34.5 dBm. Kerr combs are generated by fine adjustment of the pump wavelength. Peaks of the Cherenkov radiation are marked by the grey arrows. c, Gate voltage tunes not only the primary comb line locations (blue circles) but also the full comb bandwidth (red diamonds).

when we tune $|E_{\rm F}|$ from 0.45 eV to 0.65 eV, the effective refractive index $n_{\rm eff}$ is controlled from 1.789 + 0.058i to 1.781 + 0.001i. Figure 2c shows the measured transmission and free spectral range (FSR; the wavelength spacing between successive maxima) dependences of the GMR, at different gate voltages. In this measurement, a broadband tunable laser serves as the light source at less than 10 mW, below the comb generation threshold. For a selected resonance around 1,600 nm, when $V_{\rm G}$ is tuned from 0 V to -2 V, the extinction ratio increases from 63% to 84%, and the resonance linewidth decreases from 3.1 pm to 1.6 pm. The mode deviation from equidistance, $D_{\rm FSR} = -\beta_2 c (2\pi f_{\rm FSR})^2/n_{\rm effs}$ is 320 kHz per mode for $V_{\rm G} \approx -1$ V (anomalous dispersion) but -45 kHz per mode for $V_{\rm G} \approx -1.8$ V (normal dispersion), where c is the light velocity in vacuum and $f_{\rm FSR}$ is the frequency range of the FSR¹³. More details are shown in Extended Data Fig. 1.

Figure 2d shows the gate-tuning performance of the GMR. When the gate voltage is between 0 V and -2 V, the Fermi level remains higher than 0.4 eV, and thus graphene linear absorption in our working spectral range around 1,600 nm is strongly inhibited by Pauli blocking. As a result, the loaded Q factor of the GMR increases from about 6×10^5 to 10^6 , enabling comb generation under a 1-W pump, which is critical for both protecting the graphene monolayer from damage and stabilizing the frequency combs. We also note that Q-factor deterioration is induced by both the etching process and the linear absorption of the graphene heterostructure (Extended Data Fig. 2). For applications that require a higher Q factor, other 2D materials such as transition metal dichalcogenides with intrinsic bandgaps (for example WSe₂) could be used to construct the heterogeneous microcavities²⁹. Simultaneously, the dispersion of the resonator is dynamically tuned, varying continuously from $-62 \text{ fs}^2 \text{ mm}^{-1}$ anomalous dispersion to $+9 \text{ fs}^2 \text{ mm}^{-1}$ normal dispersion. The group velocity dispersion tuning mainly results from

d, Frequency spacing between the continuous-wave pump and the Cherenkov radiation, which is proportional to β_2/β_3 . **e**, 3-dB modulation bandwidths of 80 kHz, 200 kHz and 600 kHz are demonstrated by using optical filters with passband bandwidths of 50 nm, 9 nm and 2 nm respectively. The modulation speed is currently bounded by the ion-gel capacitance. **f**, Statistical distribution of the measured soliton states, with the same experimental parameters except for $V_{\rm G}$ which is tuned from -1.1 V to -1.6 V.

the graphene's Dirac–Fermi dynamics³⁰, with smaller contributions from the ion transport and thermal effects in the ion gel.

Next, we pump the GMR with 2-W continuous-wave laser power, with the primary comb lines (the strongest frequency combs generated from modulation instability initiation) shown in Fig. 3a under different gate voltages. For applied $V_G = -1 V$, -1.2 V and -1.5 V, the frequency offsets between the primary comb line and the pump Δf_{pri} , proportional to $(1/\beta_2)^{1/2}$, are observed at 2.36 THz, 3.25 THz and 7.17 THz, respectively. When $V_{\rm G} = -1.8$ V, the group velocity dispersion of the GMR becomes positive and hence it becomes harder to phase-match without local mode-crossing-induced dispersion. Figure 3b shows the optical spectra under carefully controlled laser-cavity detuning. In particular, at $V_{\rm G} = -1$ V, $\beta_2 \approx -62$ fs² mm⁻¹ and $\beta_3 \approx -9$ fs³ mm⁻¹ the Kerr comb has a span of about 350 nm, with highly symmetrical shape. Interestingly, with $V_{\rm G} = -1.2$ V, $\beta_2 \approx -33$ fs² mm⁻¹ and $\beta_3 \approx -630 \, \text{fs}^3 \, \text{mm}^{-1}$, we observe a frequency comb spectrum spanning 600 nm, consistent with the general route of a smaller group velocity dispersion bringing about a broader comb spectrum. The comb spectrum is highly asymmetric, with the red-side comb line intensity contributions from Cherenkov radiation. The spectral peak of the Cherenkov radiation is determined by $3\beta_2/\beta_3$, matching the measurement results. In addition, such soliton perturbation and energy transfer can be used to stabilize the Kerr frequency comb¹³. When $V_{\rm G} = -1.5$ V, $\beta_2 \approx -8 \text{ fs}^2 \text{ mm}^{-1}$ while $\beta_3 \approx -213 \text{ fs}^3 \text{ mm}^{-1}$. Because β_2 here is small (less than $10 \text{ fs}^2 \text{ mm}^{-1}$), it does not support a stable Kerr comb, the observed comb lines are not even, and the Cherenkov peak in the spectrum is also indistinguishable.

Figure 3c summarizes the gate tunability of the graphene Kerr combs, with $V_{\rm G}$ from 0 V to -2 V. For primary comb lines, their relative spectral location $\Delta f_{\rm pri} = |f_{\rm pri} - f_{\rm pump}|$ is strongly controlled, moving from 2.3 THz to 7.2 THz as $V_{\rm G}$ only changes from -1.0 V to -1.5 V. This modulation



Fig. 4 | Soliton crystals of the gated graphene-nitride microresonator. a, d, Soliton state with crystal-like defects including the single-soliton defect in a. b, c, Periodic soliton crystal states with equally spaced soliton pulses. Panels a to d are achieved with gate voltages $V_{\rm G}$ tuned at different values, ranging from -1.2 V in a to -1.5 V in d. Left panels: measured intensity transmission, illustrating the characteristic 'steps' associated

with soliton formation. Middle panels: corresponding optical spectra measurements. The pump locations are marked by black dashed lines and the Cherenkov radiation peaks are marked by grey arrows. Right panels: frequency-resolved second-harmonic autocorrelation maps of the soliton pulses. Here the grey curves show the real-time autocorrelation intensity traces.

is also influenced by the slight nonlinearity enhancement introduced by the graphene. For the full-span combs generated without missing comb lines, we also demonstrate electric-field control of their spectral span, from 38 THz to 82 THz with $V_{\rm G}$ from -1.0 V to -1.3 V (for this device, $|V_{\rm G}| > 1.3$ V does not show a good coherent comb state). Moreover, we note that the gate-tuning changes the FSRs of the combs, from 89.6 GHz at -1.0 V to 89.9 GHz at -1.5 V. Such optoelectronic tunability enables different Kerr frequency combs with a variety of properties to exist in the same device. Figure 3d next illustrates the measured locations of the Cherenkov radiation peaks in comparison with the computed designs. In contrast to the primary comb lines, the third-order dispersion plays an important role in the Cherenkov radiation. We observed three Cherenkov peaks in the window from 1,400 nm to 2,000 nm, with spectral locations $\Delta f_c = |f_c - f_{pump}|$ at values of 26.3 THz ($V_G = -1.2$ V), 49.2 THz ($V_{\rm G} = -1.3$ V) and 17.7 THz ($V_{\rm G} = -1.5$ V). The measured results well match the analytic calculation. In Fig. 3c and d, results are collected in the region of -0.4 V to -1.6 V, because when V_G is more than -0.4 V, the Q factor of the GMR is too low for comb generation; and when $V_{\rm G}$ is less than -1.6 V, the group velocity dispersion is too small to ensure a stable comb.

We estimate the modulation speed of the GMR in Fig. 3e. With $V_{\rm G}$ tuning, the output comb line intensity within the filter window is modulated temporally. The modulation speed here is bounded by ion diffusion in the heterostructure, large ion-gel capacitance on the graphene, and the optical filter bandwidth. In our current proof-of-principle demonstration, to ensure that $|E_{\rm F}|$ is sufficiently high, we use the ion-gel-based capacitor, the large capacitance (7.2 μ F cm⁻²) and slow ion diffusion (about 10⁻¹⁰ m² s⁻¹) of which limit the charge–discharge operation speed to less than hundreds of kilohertz. The optical filter bandwidth can be narrowed to improve the detection rate of the modulation by almost 7.5 times. In Fig. 3e, we show the modulated signal-to-noise ratio with a radiofrequency spectrum analyser, by

using optical filters with passband widths of 50 nm, 9 nm and 2 nm, respectively. Their corresponding bandwidths are 80 kHz, 200 kHz and 600 kHz (Extended Data Fig. 3). Although sub-megahertz modulation for the primary comb is successfully demonstrated, we note that fast modulation while preserving the full-grown Kerr comb across the entire modulation cycle could be much more challenging: with V_G tuning, not only the group velocity dispersion but also the FSR of the GMR is tuned. Compared with the primary combs shown in Fig. 3a, phase-matching of the full combs in Fig. 3b is much more sensitive: a slight variation in the FSR from the gate modulation may cause the Kerr comb to collapse. To achieve reliable, fast on–off switching in full-generated Kerr combs, inverse FSR compensation (for example, by temperature feedback) should be applied. Such sub-megahertz tunability for a Kerr comb could potentially be used in applications³¹ such as precision measurements.

Dispersion is one of the most critical cavity parameters that defines the Kerr frequency comb dynamics. The broadband dispersion modulation controlled by the gate voltage of the graphene–nitride microresonator opens up the possibility of dynamically selecting the formation path of dissipative Kerr solitons and frequency combs. By using the gate-tunable GMRs, we can engineer the dispersion dynamically to form different soliton states through electrical control. With a fixed pump power of 2 W, Fig. 3f counts the soliton states achieved in measurements for gate voltages in the range -1.6 V to -1.1 V, with the experimental conditions otherwise kept constant. In total, we have found soliton states with soliton numbers of 12, 11, 9, 8, 6, 5 and 4. More theoretical calculations and simulations are discussed in Supplementary Information section 1.4.

Figure 4 demonstrates four specific examples of soliton crystal states, under optimized gate voltages. Here the left panels show the measured intensity transmission, the middle panels demonstrate the optical spectra, and the right panels illustrate the frame-by-frame

frequency-resolved second-harmonic autocorrelation maps. These soliton states with low radiofrequency noise are achieved following Turing patterns and chaotic states before transition into the soliton states (Extended Data Fig. 4). This is characterized by a transmission step, by tuning the pump laser gradually into the cavity resonance. Figure 4a shows two examples of the soliton state with missing pulses, at a gate voltage of -1.2 V. The corresponding pump laser wavelength is around 1,600.2 nm. The optical spectra of these states are characterized by the apparent existence of groups of comb lines that are separated by multiple cavity FSRs. Within each comb group, weaker single-FSR comb lines are present, and they effectively connect all comb groups without any spectral gaps. For the examples shown in Fig. 4a and d, the comb groups are separated by 8 FSR, 5 FSR and 12 FSR respectively. In the time domain, the autocorrelation traces reveal the common features of missing pulses in the otherwise equally spaced soliton states with higher effective repetition rate. The self-organization of multiple soliton pulses into a train of equally spaced pulses resembles the crystallization process and is therefore termed a soliton crystal³², and the missing pulse structure is analogous to defects in crystal lattices. Our graphene-nitride heterogeneous microresonator thus provides a platform for study of soliton physics that is tunable through the gate-voltage and Fermi level. We also note that when the soliton crystals are formed, the emitted soliton Cherenkov radiations are sharp and narrow, as marked by the grey arrows in Fig. 4.

Soliton crystals are formed because of the strong mode interaction and intracavity interferences, and thus their evolution dynamics depend critically on the exact dispersion profile of the microresonator. By further optimizing the group velocity dispersion and third-order dispersion through gate tuning, we demonstrate two periodic soliton crystal states. Figure 4b shows a four-soliton state with $V_{\rm G} = -1.3 \,\rm V$ and pump laser at approximately 1,584.2 nm, while Fig. 4c shows a 11-soliton state with $V_{\rm G} = -1.4 \,\rm V$ and pump laser at approximately 1,600.1 nm. Intriguingly, these soliton crystal states show remarkable stability, and they can robustly survive a pump power fluctuation up to $\pm 2 \, dB$, or wavelength offset up to $\pm 300 \, pm$. The soliton crystal formation is also akin to harmonic mode-locking in which a stable highrepetition-rate pulse train can be attained even in longer cavities, and it is of interest in applications such as high-speed communication, comb spectroscopy and data storage. This realization of a charge-tunable graphene heterostructure for controllable frequency combs and soliton dynamics opens a new architecture at the interface of single-atomiclayer nanoscience and ultrafast optoelectronics.

Online content

Any Methods, including any statements of data availability and Nature Research reporting summaries, along with any additional references and Source Data files, are available in the online version of the paper at https://doi.org/10.1038/s41586-018-0216-x.

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Additional information

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METHODS

Theoretical analysis. The refractive index of graphene, n_G , is determined by its permittivity ε_G as $n_G = \varepsilon_G^{1/2}$, where $\varepsilon_G = \{-\text{Im}(\sigma_G) + i\text{Re}(\sigma_G)\}/\{2\pi f \Delta\}$ (ref. ²⁰); here σ_G is the conductivity of graphene, f is the optical frequency, and $\Delta = 0.4$ nm is the thickness of the graphene monolayer. Particularly, $\partial^n \text{Re}(n_G)/\partial \lambda^n$ determines the *n*th-order dispersion, while $\text{Im}(n_G)$ determines the waveguide loss. In Supplementary Information section 1.1, we describe in more detail how the transmission of graphene is determined by its quasi Fermi level E_F . By gating graphene via an external field, one can conveniently control both the group velocity dispersion β_2 and the third-order dispersion β_3 of a graphene monolayer. Kerr comb generation in the GMR. In Supplementary Information section 1.2, we analyse the comb formation dynamics by LLE modelling. In Supplementary Information section 1.3, we describe the third-order nonlinearity of graphene. In Supplementary Information section 1.4, we provide detailed simulations of the soliton generations in the GMR.

Device design and fabrication. First, in a silicon foundry, we nanofabricated a high-Q silicon nitride microresonator with measured loaded $Q \approx 1.6 \times 10^6$ (intrinsic $Q \approx 1.8 \times 10^6$) and FSR ≈ 90 GHz in a 350-µm-diameter ring structure. The nitride core has a $1,200 \times 800 \text{ nm}^2$ cross-section, a 600 nm gap to the input-output coupling waveguide of $1,000 \times 800 \text{ nm}^2$ cross-section, and a top oxide cladding. Next, single-atomic-layer graphene was grown using a chemical vapour deposition method and transferred onto the exposed region of the nitride ring (with etched SiO₂ window). The monolayer graphene was then lithographically patterned and oxygen plasma etched into an $80\,\mu\text{m} \times 100\,\mu\text{m}$ sheet. Metallization of sourcedrain electrodes was achieved through standard photolithography, followed by electron-beam evaporation of Ti/Au (20/50 nm thick). Here the electrode pad size was $80 \mu m \times 60 \mu m$. Subsequently, we integrated an ionic liquid (DEME-TFSI, N,Ndiethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl)imide) as the gate dielectric, resulting in an electric double-layer graphene transistor. More details are shown in Supplementary Information section 2 and Supplementary Fig. 12.

Experimental set-ups. We implemented a temperature-controlled optical set-up for the frequency comb generation. The spectral tunable range of our drive laser is 1,480 nm to 1,640 nm, and the maximum output power of our erbium-doped fibre amplifier (EDFA, BKtel) in the L-band is 3.16 W (35 dBm). The GMR transmission is measured by using the same tunable laser, swept through its full wavelength tuning range at a speed of 40 nm s^{-1} , to obtain dispersion and Q factors. A fibre-coupled hydrogen cyanide gas cell (HCN-13-100, Wavelength References Inc.) and an unbalanced fibre Mach-Zehnder interferometer are used for calibration. To measure the stability and soliton states of our frequency comb, heterodyne and autocorrelation measurements are implemented. For the heterodyne measurement, a stable continuous-wave laser with narrow linewidth (300 kHz, New Focus) is applied as the heterodyne reference for the beat notes. For the autocorrelation measurement, a fibre with zero group velocity dispersion, made of a 7-m dispersion-compensating fibre and a 15-m single-mode fibre, is used to guide the microresonator output to the autocorrelation set-up with minimal pulse broadening and distortion. More details about the experimental set-ups are shown in Supplementary Figs. 13-16.

Soliton-step evolution process in the graphene frequency comb. Complementary to Fig. 4, Extended Data Fig. 4 illustrates the soliton states at a gate voltage of -1.2 V, for different laser-cavity detunings. With the simultaneous optical and radiofrequency spectra measurements, the frequency comb initiates from the Turing pattern (state i) into the high-noise patterns (states ii and iii, with sub-comb competition) before settling down into the low-noise soliton comb (states iv and v). With further detuning, the soliton comb goes back into the high-noise regime (state vi), owing to the thermal instability in the cavity. We then beat the comb lines of the soliton state (state iv) and the four-soliton state (state v). Here, the 9th mode and the 56th mode denote the offsets from the pump line. The beat notes show an intensity contrast ratio of more than 40 dB, with a linewidth of 200 kHz, verifying that there is clean and stable soliton generation.

Data availability. The data that support the findings of this study are available from the corresponding authors on reasonable request.





Extended Data Fig. 1 | **Measured gate-tunable coupling and dispersions** in a GMR. a, Dips at approximately 1,600 nm, with different $V_{\rm G}$. b, Correlation of the round-trip transmissions and the bus transmissions for the resonator, obeying $T = (\alpha - |t|)^2 / (\alpha - \alpha |t|)^2$. Here, $1 - \alpha$ is the cavity loss per round trip, and 1 - t is the bus-to-cavity coupling rate. In our

experiment, the graphene ring resonator is under-coupled originally, as the blue dot shows. **c**, Group velocity dispersion in range of 1,500 nm to 1,700 nm. Here, the curves show the calculated results, while dots show measured data. **d**, Calculated third-order dispersion in range of 1,500 nm to 1,700 nm.

LETTER RESEARCH



Extended Data Fig. 2 | Comparative optical transmissions of the heterogeneous graphene-nitride ring. a, Spectral transmission of the silicon nitride ring resonator under the silica overcladding. b, Spectral transmission of the silicon nitride ring resonator after buffer-oxide etching to remove the silica overcladding. c, Spectral transmission of the graphene/

ion-gel-based nitride ring resonator, heavily p-doped ($V_{\rm G} = -2$ V). d, Loaded Q factor around 1,600 nm. e, FSR, which is sensitive to the geometry modification. f, Mode non-equidistances, D_2 . d and e are measured at $\lambda = 1,600$ nm. In this figure, the error bars denote the typical system error.

RESEARCH LETTER



Extended Data Fig. 3 | **An implementation of the graphene primary frequency comb gate-modulation. a**, Method for measuring the modulated comb. Keeping bias $V_{\rm G} = -1.2$ V, we control the laser-cavity detuning to generate a primary comb such as the grey spectrum shown here. To filter off the 1,600-nm continuous-wave pump, we apply a C-band filter, selecting the comb lines in the C-band only. A signal generator (maximum amplitude of 2 V, HP3312) is applied to modulate the gate

voltage between -1.2 V and -1.8 V. In this process, primary comb lines in the filter window are modulated by the gate signal; the modulation is monitored by using an oscilloscope (500 MHz, Rigol DS1054) and an electrical spectrum analyser (ESA, 3 GHz, Agilent CXA9000A). **b**, Examples of radiofrequency spectra of the modulated combs, filtered by an optical filter (1,530 nm to 1,570 nm).

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Extended Data Fig. 4 | Example measurements of the graphene soliton comb formation. a, Under $V_{\rm G} = -1.2$ V (Fermi level 0.59 eV), when the wavelength of the pump ($\lambda_{\rm p}$) is tuned from 1,600.00 nm to 1,600.23 nm, the Kerr frequency comb is generated gradually. When $\lambda_{\rm p}$ is tuned between 1,600.15 nm and 1,600.19 nm, two multi-soliton states with low phase noise are achieved (states iv and v). b, Corresponding radiofrequency (RF) amplitude noise of the six states. In **a** and **b**, the pump power is kept at

34.5 dBm. Cherenkov radiation of the multi-soliton comb is narrow and sharp. **c**, Zoom-in of the eight-soliton crystal spectrum. The FSR changes from 89 GHz to 718 GHz, owing to the soliton-crystal-based longitude mode interaction. **d**, Beat note for the comb lines of the eight-soliton state (red; ninth comb line offset from the pump) and the four-soliton state (green; 56th comb line offset from the pump).