Aluminum nitride nanophotonics for beyond-octave soliton microcomb generation and self-referencing

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Frequency microcombs, alternative to mode-locked laser and fiber combs, enable miniature rulers of light for applications including precision metrology, molecular fingerprinting and exoplanet discoveries. To enable frequency ruling functions, microcombs must be stabilized by locking their carrier-envelope offset frequency. So far, the microcomb stabilization remains compounded by the elaborate optics external to the chip, thus evading its scaling benefit. To address this challenge, here we demonstrate a nanophotonic chip solution based on aluminum nitride thin films, which simultaneously offer optical Kerr nonlinearity for generating octave soliton combs and quadratic nonlinearity for enabling heterodyne detection of the offset frequency. The agile dispersion control of crystalline aluminum nitride photonics permits high-fidelity generation of solitons with features including 1.5-octave spectral span, dual dispersive waves, and sub-terahertz repetition rates down to 220 gigahertz. These attractive characteristics, aided by on-chip phase-matched aluminum nitride waveguides, allow the full determination of the offset frequency. Our proof-of-principle demonstration represents an important milestone towards fully integrated self-locked microcombs for portable optical atomic clocks and frequency synthesizers.

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Optical frequency combs, developed from solid-state or fiber mode-locked lasers, have evolved into photonic chip-based sources that feature the potential towards a miniaturized footprint and reduced cost. Among various chip-scale schemes, microresonator Kerr frequency combs ("microcombs" hereafter) are of particular interest because of their high scalability for photonic integration. To enable phase-coherent microcombs, substantial efforts have been made towards soliton mode-locking on the one hand, and unveiling rich soliton physics on the other hand. Specifically, octave-spanning soliton microcombs are important because it permits phase locking of the carrier-envelope offset (CEO) frequency ($f_{\text{CEO}}$) via well-known $f$-$2f$ interferometer, and are prerequisite for chip-scale implementation of precision metrology, frequency synthesizers and optical clocks. To date, silicon nitride ($\text{Si}_3\text{N}_4$) nanophotonics has proven viable for octave soliton operations with a terahertz repetition rate ($f_{\text{rep}}$). Nevertheless, such a large $f_{\text{rep}}$ is not amenable for direct photodetection and poses challenges to access the CEO frequency with a value up to $f_{\text{rep}}$. In the meantime, the lack of intrinsic quadratic $\chi^{(2)}$ nonlinearities in $\text{Si}_3\text{N}_4$ films typically requires an external frequency doubler and off-chip optical circuity for deriving the CEO frequency. These off-chip optical components compromise the scaling advantage of microcombs and significantly set back self-locked microcombs for portable applications.

Aluminum nitride (AlN) semiconductors exhibit a noncentrosymmetric crystal structure, thereby possessing inherent optical $\chi^{(2)}$ nonlinearity as well as Pockels electro-optic and piezoelectric properties. Apart from the advances in ultraviolet light-emitting diodes and quantum emitters, AlN has also proved viable for low-loss nanophotonics in high-efficiency second-harmonic generation (SHG) and high-fidelity Kerr and Pockels soliton mode-locking. Therefore, it is feasible to establish an on-chip $f$-$2f$ interferometer provided that an octave AlN soliton microcomb is available. This is a solution that is favored here comparing with the heterogeneous integration of comb spectral extension via dispersive wave (DW) emissions. This is a solution that is favored here comparing with the heterogeneous integration. This is a solution that is favored here comparing with the heterogeneous integration.

In this article, we demonstrate high-fidelity generation of octave soliton microcombs and subsequent $f_{\text{CEO}}$ detection using AlN-based nanophotonic chips. Thanks to mature epitaxial growth, AlN thin films with highly uniform thickness are available, thus permitting lithographic control of group velocity dispersion (GVD) for comb spectral extension via dispersive wave (DW) emissions. Our octave soliton microcombs possess separate dual DWS and moderate $f_{\text{rep}}$ of 433, 360, and 220 gigahertz, and are found to be reproducible from batch-to-batch fabrications. The results then allow us to capture the $f$-$2f$ beatnote through on-chip SHG in phase-matched AlN waveguides. Our work establishes the great potential of non-centrosymmetric AlN photonic platforms for achieving portable self-locked microcomb sources in the near future.

Results

Experimental scheme description. Figure 1a illustrates the implementation of microcomb-based $f$-$2f$ interferometry from a nanophotonic chip. The strategy is to leverage non-centrosymmetric photonic media for co-integration of $\chi^{(2)}$ octave soliton microcombs and $\chi^{(2)}$ SHG doublers. For a proof-of-principle demonstration, we adopt an auxiliary laser (at $f_{\text{aux}}$) to obtain sufficient SHG power (at $2f_{\text{aux}}$) from phase-matched optical waveguides. The use of the auxiliary laser can be eliminated by exploiting microring-based architectures to boost the SHG efficiency. By subsequently beating $f_{\text{aux}}$ and $2f_{\text{aux}}$ with the $f_0$ and $2f_0$ comb lines at their corresponding beatnotes of $\delta_1$ and $\delta_2$, the $f_{\text{CEO}}$ signal reads:

$$f_{\text{CEO}} = \delta_2 - 2\delta_1$$

The AlN thin films in this work were epitaxially grown on a c-plane sapphire substrate via metal-organic chemical vapour deposition. As illustrated in Fig. 1b, the AlN epilayer exhibits a hexagonal wurtzite structure with a unit cell shown in the bottom, highlighting the non-centrosymmetry. We also show an overall 2-inch AlN-on-sapphire wafer featuring a broadband transparency and a favored film thickness (Fig. 1c) that are crucial factors to ensure octave GVD control. Great attention was also paid to the film crystal quality and surface roughness for low-loss photonic applications. The AlN nanophotonic chips were manufactured following electron-beam lithography, chloride-based dry etching, and silic on dioxide ($\text{SiO}_2$) coating processes and were subsequently cleaved to expose waveguide facets. The intrinsic optical quality factors ($Q_{\text{int}}$) of the AlN resonators were characterized to be $\sim$1–3 million depending on the waveguide geometries. The detailed film and device characterization is presented in “Methods” and Supplementary Fig. 1.

Since wurtzite AlN manifests optical anisotropy for vertically or horizontally-polarized light, we engineer the waveguide structures for optimizing the performance of fundamental transverse magnetic (TM$_{00}$) modes, which allows the harness of its largest $\chi^{(2)}$ susceptibility to ensure high-efficiency SHG. To expand microcomb spectra out of the anomalous GVD restriction, we exploit soliton-induced DW radiations by tailoring the resonator’s integrated dispersion ($D_{\text{int}}$):

$$D_{\text{int}} = -\frac{D_3}{2!} \mu^2 + \frac{D_3}{3!} \mu^3 + \sum_{i=4}^{\infty} \frac{D_i}{i!} \mu^i$$

where $D_3$, $D_5$, and $D_i$ are $i_{th}$-order GVD parameters, while $\mu$ indexes the relative azimuth mode number with respect to the pump ($\mu = 0$). In the dispersion modeling, we have accounted for both the material (AlN) and geometry (cross section, bending radius, and slanted sidewall) dispersion. To prevent avoided mode crossing from interrupting soliton mode-locking, we adopt a weak pulley-coupling configuration (concentric angle of 6°), which helps suppress the excitation of higher-order resonator modes (Supplementary Fig. 1c).

Octave soliton microcombs. Our GVD engineered AlN resonators are coated with a $\text{SiO}_2$ protection layer, making it less susceptible to the ambient compared with the air-cladded $\text{Si}_3\text{N}_4$ counterpart. An example of the resonator modal profile is shown in the inset of Fig. 2a. The top panel of Fig. 2a plots the $D_{\text{int}}$ curve from a 50 μm-radius AlN resonator through numerical simulation (see “Methods”). In spite of the limited anomalous GVD window (solid light blue region), octave microcomb operation is feasible via DW radiations at phase-matching conditions $D_{\text{int}} = 0$, allowing for spectral extension into normal GVD regimes (solid light orange regions). Note that the occurrence of such dual DWs benefits from the optimal film thickness in our AlN system, while the DW separation is agilely adjustable over one octave through the control of resonator’s dimensions (Supplementary Fig. 2). Around the telecom band, the $D_{\text{int}}$ value (red dots) was characterized by calibrating the resonator’s transmission with a fiber-based Mach-Zehnder interferometer. The experimental result matches well with the simulated one (inset of Fig. 2a) with an extracted $D_2/2\pi$ of $\sim$6.12 MHz.
We then explore soliton mode-locking based on a rapid frequency scan scheme to address the abrupt intracavity thermal variation associated with transitions into soliton states. The soliton spectrum is recorded using two grating-based optical spectrum analyzers (OSAs, coverage of 350–1750 nm and 1500–3400 nm). The experimental setup is detailed in Supplementary Fig. 4. The bottom panel of Fig. 2a plots the soliton spectrum from a 50 μm-radius AlN resonator, featuring a moderate f_{rep} of 433 GHz and an observable spectral span of 1.05–2.4 μm, exceeding one optical octave. Meanwhile, soliton-induced DW radiations occur at both ends of the spectrum, in agreement with the predicted D_{int} curves. Note that the high-frequency DW exhibits an evident blue shift from the D_{int} = 0 frequency, which is mainly ascribed to Raman-induced soliton red shifts (relative to the pump frequency). The soliton recoils make less impact here due to dual DW radiations. This conclusion is supported by our modeling when comparing the soliton spectra with and without Raman effects (Supplementary Fig. 2b).

The single crystal nature of AlN thin films permits reproducible optical refractive indices in each manufacture run. This, in combination with their uniform film thickness control, leads to a high predictability for the dispersion engineering, making it feasible to predict octave soliton combs at various repetition rates. For instance, our GVD model indicates that octave spectra with repetition rates further decreased by two times are anticipated from 100 μm-radius AlN resonators at optimal widths of 3.3–3.5 μm (Supplementary Fig. 3a). Figure 2b plots the recorded soliton comb spectrum at a resonator width of 3.5 μm, where a f_{rep} of ~220 GHz and dual DWs separated by more than one octave are achieved simultaneously. Such a low f_{rep} is amenable for direct photodetection with state-of-the-art unitravelling-carrier photodiodes. We also noticed the occurrence of a weak sharp spectrum around 130 THz, which might arise from modified local GVD due to avoided mode crossing. In our nanophotonic platform, we could further predict resonator geometries for achieving octave solitons with an electronically detectable f_{rep} of ~109 GHz (Supplementary Fig. 3b). Nonetheless, the strong competition between Kerr nonlinearities and stimulated Raman scattering (SRS) must be taken into account since the free spectral range (FSR) of the resonator is already smaller than the Δg frequency linewidth (~138 GHz) in AlN epilayers.

Since the SHG from the auxiliary laser (1940–2000 nm) available in our laboratory is beyond the soliton spectral coverage shown in Fig. 2, we further adjust the resonator dimensions for extending microcomb spectra below 1 μm. As plotted in Fig. 3a, the phase-matching condition (D_{int} = 0) for high-frequency DW radiations below 1 μm is fulfilled by elevating the resonator radius to 60 μm while maintaining its width around 2.3 μm. In the meantime, low-frequency DWs could also be expected and their spectral separation is adjustable by controlling the resonator width. Guided by the tailored D_{int} curves, we fabricated the AlN resonators and recorded octave soliton spectra at a f_{rep} of ~360 GHz (Fig. 3b). Lithographic control of DW radiations (indicated by vertical arrows) is also verified by solely adjusting the resonator width, allowing the spectral extension below 1 μm (width of 2.3 or 2.4 μm). The low- and high-frequency DWs are found to exhibit distinct frequency shifting rates, consistent with the D_{int} prediction. The observable soliton spectra (from top to bottom of Fig. 3b) cover 1.5, 1.3, and 1.2 optical octaves by normalizing the total span (Δf) to its beginning frequency (f_{0}), that is Δf/f_{0}. Such a definition permits a fair comparison among
soliton microcomb generation in distinct pump regimes across different material platforms, suggesting high competitiveness of our AlN microcomb span comparing to state-of-the-art values reported in Si$_3$N$_4$ microresonators$^{26}$.

**On-chip second harmonic generator.** We then explore the co-integration of SHG based on the $\chi^{(2)}$ susceptibility of AlN for matching the DW peak below 1 μm (middle panel of Fig. 3b). To fulfill the demanding requirement of spectral overlaps with the microcomb, we adopt a straight waveguide configuration, which allows a broader phase-matching condition albeit at the cost of reduced conversion efficiencies comparing to its counterpart using dual-resonant microresonators$^{34,50}$. Through modeling, we predict an optimal waveguide width of ~1.38 μm for fulfilling the modal-phase-matching condition (Supplementary Fig. 5a), while the actual waveguide width was lithographically stepped from 1.32 to 1.46 μm (spacing of 5 nm) accounting for possible deviations during the manufacturing process.

Figure 4a shows a section of 6 cm-long SHG waveguides co-fabricated with the microcomb generator. At a fixed fundamental wavelength (1970 nm), we located the phase-matching waveguide wavelength (1990 nm). The resonator inset indicates a large 3-dB phase-matching bandwidth of ~0.8 nm, which, together with an external heater for thermal fine-tuning, is sufficient to cover the target comb lines for subsequent heterodyne beating.

**Nanophotonics-based $f$–$2f$ interferometry.** By combining outgoing light from optimal AlN soliton and SHG generators on the calibrated OSAs, we are able to estimate the $f$–$2f$ beatnote frequency to be approximately 32 GHz limited by the resolution of the OSAs. To electronically access the $f_{\text{CEO}}$ signal in real time, we employ a scheme sketched in Fig. 5a. The recorded soliton spectrum after suppressing pump light by a fiber Bragg grating (FBG) indicates a high off-chip power close to ~40 dBm for the high-frequency DW (Supplementary Fig. 6a). Meanwhile, a wavelength-division multiplexer (WDM) is utilized to separate the $f$ and $2f$ frequency components before sent into the photodetectors (PDs). Two tunable radio frequency (RF) synthesizers are introduced as the local oscillators (LO1 and LO2) to down convert the photodetector signals for effective capture of the $f$–$2f$ beat signal at a convenient low-frequency band with an electronic spectrum analyzer (ESA, range of 20 Hz–26.5 GHz).

As highlighted in Fig. 5b, we record two down-converted beatnotes of $\Delta f_1$ and $\Delta f_2$ with a signal-to-noise ratio (SNR) of 10 dB at a resolution bandwidth of 1 MHz. A much higher SNR is anticipated by applying a finer detection bandwidth upon locking the telecom pump laser as well as the $f_{\text{CEO}}$ frequency$^{23,24}$. The corresponding $f$–$2f$ beatnote is $f_{\text{CEO}} = 2f_{\text{LO1}} + f_{\text{LO2}} - \Delta f_2$. 

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**Fig. 2 Octave soliton microcombs at hundreds of gigahertz repetition rates.** a Top: integrated dispersion ($D_{\text{int}}$) of a 50 μm-radius (R) AlN resonator (cross section: 1.0 × 2.3 μm$^2$), where the anomalous and normal group velocity dispersion (GVD) regimes are indicated by solid light blue and orange colors in the bottom. Insets: zoom-in view of measured (red dots) and simulated (blue curve) $D_{\text{int}}$ values in the center, and electric field $\text{abs}(E_z)$ of the TM$_{00}$ resonator mode with a normalized color bar in the right. Bottom: soliton microcomb spectra from the experiment (Exp., blue curve) and simulation (Sim., red curve) at an on-chip pump power of ~390 mW. The resonator $Q_{\text{int}}$ is 1.6 million and the repetition rate ($f_{\text{rep}}$) is estimated to be around 433 GHz. b Soliton microcomb spectrum from a 100 μm-radius AlN resonator (cross section: 1.0 × 3.5 μm$^2$) with a decreased $f_{\text{rep}}$ of ~220 GHz. The applied pump power is -1W at a resonator $Q_{\text{int}}$ of 2.5 million.
since the local oscillator frequencies $f_{LO1}$ and $f_{LO2}$ are chosen to be larger than beatnotes of $\delta_1$ and $\delta_2$. Based on the relative frequency positions of the auxiliary laser and its neighboring comb tooth (Supplementary Fig. 6b), we reach an actual $f_{CEO} = FSR - f_{CEO}$ due to the involvement of $f_n$ and $2f_{n+1}$ comb lines in the heterodyne beating. On the other hand, the $f_{LO1}$ and $f_{LO2}$ frequencies are freely adjustable up to 40 and 20 GHz in our scheme, which could further expand the accessible range of the $f_{CEO}$ frequency based on the down-conversion process presented here. Meanwhile, the RF synthesizers are synchronized to a common external frequency reference, suggesting that the captured down-converted $f-2f$ signals are available for further locking the comb teeth in a feedback loop.

\[ D_{int} \]

\[ f_{rep} \]

\[ R = 60 \mu m \]

\[ W = 2.3 \mu m \]

\[ W = 2.4 \mu m \]

\[ W = 2.5 \mu m \]

\[ 120 \quad 140 \quad 160 \quad 180 \quad 200 \quad 220 \quad 240 \quad 260 \quad 280 \quad 300 \quad 320 \]

\[ \text{Frequency (THz)} \]

\[ \text{Power (dBm)} \]

\[ \text{Wavelength (nm)} \]

\[ \text{Data Sinc fit} \]

\[ \text{Shg power (W)} \]

\[ \text{Ey (a.u.)} \]

\[ \text{TM20 TM00} \]

\[ 600 \mu m \]

\[ \text{60 dB div}^{-1} \]

\[ 2.3 \mu m \]

\[ 2.5 \mu m \]

\[ \text{SHG power (W)} \]

\[ \text{Ey (a.u.)} \]
wafer, we can reliably locate the desired region for reproducible octave device fabrication.

To further reduce the propagation loss, the AlN photonic chips were annealed at 1000 °C for 2 h. The resonator Q-factors were probed by sweeping a tunable laser (Santec TSL-710) across the cavity resonances and then fitted by a Lorentzian function. In the 100-μm-radius AlN resonators (width of 3.5 μm), we achieve a recorded Qint of 3.0 million, while the 50-μm-radius resonators (width of 2.3 μm) exhibit a decreased Qint of 1.6 million, indicating the dominant sidewall scattering loss of our current fabrication technology. The related resonance curves are plotted in Supplementary Fig. 1c and d.

Numerical simulation. The Dint of the AlN resonators is investigated using a finite element method (FEM) by simultaneously accounting for the material and geometric chromatic dispersion. The overall Dint value is approximated with a fifth-order polynomial fit applied to the simulated modal angular frequencies: \( \omega_m = \omega_p + \mu D + D_{int} \), where \( D_{1/2} \) is the resonator’s FSR at the pump mode \( \mu = 0 \).

The spectrum width of octave soliton microcombs is numerically explored based on nonlinear coupled mode equations by incorporating the Raman effect:\( \chi^{(3)} \)

\[
\frac{\partial}{\partial t} \psi = - \left( \frac{\gamma}{2} + i \Delta_k \right) \psi + i g_{\text{eff}} \sum_{k,n} \hat{a}_k \hat{a}_n \psi(1 + n - k - \mu)
- i g_{\text{eff}} \sum_{k,n} \hat{a}_k \hat{R} \sum_{l} \psi(1 + k - l - \mu) + \Delta_k \psi
\]

Here \( a \) and \( R \) are the mode amplitudes of cavity photons and Raman phonons with subscripts \( k, l, n \) being the mode indices, while \( g_{\text{eff}} \) represent the nonlinear coupling strength of Kerr and Raman processes, respectively. The driving signal strength is \( \xi = \mu \sqrt{\frac{\omega_m}{\gamma}} \) at an on-chip pump power \( P_{\text{pump}} \). \( \omega_m (\epsilon_m) \) denotes the total (external) cavity decay rate of the \( \mu \)th photon mode, and \( \omega_p \) is the Raman phonon decay rate. The detuning from a \( D_{1/2} \)-spaced frequency grid is indicated by \( \Delta_k = \omega_m - \omega_p - D_{1/2} \) and \( \Delta_k = \omega_m - \mu D_{int} \). The pump \( \omega_p \) being and Raman shift angular frequencies, respectively.

In the simulation, we set the time derivative of Raman items in Eq. (4) to zero to speed up the computation since the decay rate of phonons is much larger than that of photons. We also consider frequency-independent \( \epsilon_m(\omega_m) = 120 \text{ MHz} \) and \( \omega_p(\omega_p) = 75 \text{ MHz} \) based on measured Q-factors of 50-μm-radius AlN resonators (Supplementary Fig. 1c). Because incident light is TM-polarized, the involved \( \chi^{(3)} \) Raman phonon in AlN exhibits an \( \omega_p(\omega_p) = 18.3 \text{ THz} \) with an FWHM of \( \omega_p(\omega_p) = 138 \text{ GHz} \). The \( g_{\text{eff}} \) is calculated to be 0.73 Hz for a given nonlinear refractive index \( n_2 = 2.3 \times 10^{-19} \text{ m}^2/\text{W} \), while an optimal \( g_{\text{eff}} \) for 2 = 0.29 MHz is adopted, resulting in a soliton spectrum matching well with the measured one in Fig. 2a. The simulated high frequency DW also exhibits an evident blue shift comparing with the case of \( g_{\text{eff}} = 0 \text{ MHz} \) (Supplementary Fig. 2b).

Data availability

The spectral data that support the findings of this study are available in figshare repository: https://doi.org/10.6084/m9.figshare.15167895v1.

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Discussion

We demonstrate nanophotonics-based implementation of \( f-2f \) interferometry by leveraging \( \chi^{(3)} \) octave solitons and \( \chi^{(3)} \) SHG co-fabricated from a non-centrosymmetric AlN photonic platform. Thanks to agile GVD engineering offered by epitaxial AlN thin films, our octave soliton microcombs can be reliably produced with dual DWs and sub-THz repetition rates (220–433 GHz) that are accessible with unit travelling-carrier photodiodes. The overall soliton spectral span is adjustable up to 1.5 octaves, on a par with state-of-the-art values (1.4 octaves) reported in Si3N4 microsoliton spectral span is adjustable up to 1.5 octaves, on a par with waveguides, thus allowing for spectral overlap with the desired experimental details are introduced in Supplementary Fig. 6c.

Methods

Nanofabrication. The surface roughness and crystal quality of our AlN epilayer were respectively characterized by an atomic force microscope and an X-ray diffraction scan, indicating a root-mean-square roughness of 0.2 nm in 1 × 1 μm² region and an FWHM linewidth of ~46 and 1000 arcsec along [002] and [102] crystal orientations, respectively. The film thickness was mapped by a spectroscopic ellipsometer (J.A. Woollam M-2000), providing a quick and preliminary selection of the desired AlN piece for octave soliton generation with dual DWs. As shown in Supplementary Fig. 1a, in spite of varied film thicknesses across a 2-inch AlN
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