

Frequency-agile integrated mid-infrared electro-optic comb

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Frequency-agile integrated mid-infrared electro-optic comb

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Abstract

Mid-infrared frequency combs provide highly sensitive and selective access to molecular vibrational transitions, making them attractive for integrated spectroscopic sensing. However, compact sources offering broad wavelength coverage together with independent electronic control of comb parameters remain challenging to realize. Here, we demonstrate mid-infrared electro-optic frequency-comb generation through difference-frequency generation in periodically poled thin-film lithium niobate waveguides implemented on two substrates, namely silicon dioxide and sapphire. Our architecture combines a continuous-wave near-infrared pump with a telecom-band electro-optic comb operated in either single- or dual-comb configurations, generated on-chip. Difference-frequency generation allows downconversion of the telecom comb to the mid-infrared. The comb spacing is defined by the drive radio frequency, while the comb center frequency is independently controlled by the telecom seed wavelength. The quasi-phase-matching condition can be further tuned through temperature control. We generate mid-infrared combs with an instantaneous bandwidth of approximately 6 nm and a center-wavelength tuning range exceeding 200 nm on a single device. A direct comparison of the device on two substrates reveals the advantage of lithium niobate on sapphire for operation beyond 3.5 μm , providing higher output power, improved conversion efficiency, and extended spectral coverage up to 4.5 μm . Finally, we demonstrate on-chip dual-comb generation in the mid-infrared with a tunable relative frequency offset between the two combs. Low-frequency beat notes between corresponding comb lines are detected over a 140 nm MIR wavelength range. This unique integrated mid-infrared electro-optic comb platform combines independent electronic control of the comb center frequency, line spacing, and frequency offset with on-chip dual-comb generation.

1 Main

The mid-infrared (MIR) spectral range, spanning approximately 3-14 μm , offers major benefits for free-space optical applications, including high atmospheric transparency, reduced scattering from micrometer-scale aerosols, and improved resilience to turbulence [1–6]. In addition, many environmentally and biologically relevant molecules exhibit strong and distinctive vibrational absorption lines in this range, often significantly stronger than in the near-infrared (NIR), allowing highly sensitive and selective spectroscopic detection [7, 8]. However, realizing the full potential of this spectral range requires source technologies capable of combining broadband wavelength coverage and high spectral purity, a combination that has proven challenging to demonstrate efficiently on integrated platform. Optical frequency combs have emerged as a particularly well-suited tool to meet this need, providing broadband, evenly spaced, and phase-coherent spectral lines that act as optical frequency rulers. In the context of MIR spectroscopy, this capability enables dual-comb spectroscopy [9], high-resolution metrology [10], and rapid, parallel detection of multiple absorption features with high sensitivity and spectral resolution [11, 12]. A wide range of approaches to frequency-comb generation has been developed, most prominently in the visible and NIR spectral regions [13]. Among these, mode-locked lasers represent a mature and well-established platform reaching industry implementation [14]. In parallel, Kerr nonlinear microresonators have emerged as a powerful alternative, capable of generating octave-spanning spectra and achieving outstanding performance in the NIR [15–17] but suffering from instability. Electro-optic (EO) comb generators provide another approach, in which a continuous-wave laser is modulated using radio-frequency (RF)-driven electro-optic phase modulators. This technique enables combs with excellent stability and tunable comb spacing. To date, many integrated EO comb demonstrations have relied on lithium niobate (LiNbO_3 , LN), owing to its low propagation loss, broad transparency window extending beyond 4.5 μm , and strong second-order nonlinearity $\chi^{(2)}$, with a Pockels coefficient of up to 33 pm/V [18]. In particular, the thin-film lithium niobate on insulator (LNOI) platform enables strong confinement and compact device architectures. As a result, LNOI has driven rapid progress in electro-optic devices, with demonstrated modulation bandwidths exceeding 100 GHz and with a modulation efficiency below 1 V $\cdot$ cm [19, 20]. At 1.5 μm , non-resonant LNOI modulators have achieved EO comb bandwidths of 20 nm with 25 GHz line spacing [21]. In contrast, resonant ring-based devices have demonstrated significantly broader bandwidths of up to 132 nm with 30.925 GHz spacing and conversion efficiencies approaching 30% [22–24]. More recently, resonant EO comb generation has also been demonstrated in alternative material platforms like lithium tantalate [25]. Although the achievable bandwidth of EO combs is generally limited by modulator efficiency, dispersion, and available RF drive power, they still provide a highly coherent and scalable route that is particularly attractive for integrated photonics. Nevertheless, despite rapid progress in integrated MIR modulators [26, 27], extending electro-optic comb generation to the MIR remains challenging. This is primarily due to the limited availability of efficient phase modulators at these wavelengths, which typically exhibit large $V_\pi L$ products because of weaker optical confinement and the relative immaturity of integrated MIR photonic platforms. Efficient integrated MIR EO-comb generation critically requires a low $V_\pi L$

product [28]. Notably, MIR modulators on lithium niobate on sapphire (LNOS) have been demonstrated [28] to avoid the strong absorption of the silicon dioxide layer in conventional LNOI above $3.5\text{ }\mu\text{m}$ [29]. Low- V_π MIR modulators based on suspended LNOI waveguides have also recently enabled broadband comb generation; however, this approach entails increased fabrication complexity and susceptibility to mechanical damage [30].

Quantum cascade laser (QCL) combs are attractive owing to large bandwidth and high available power per line. Several studies have demonstrated MIR comb generation through intra-cavity phase modulation using QCLs [31], including quantum-walk QCL combs that rely purely on phase modulation [32]. More recently, racetrack QCLs have been shown to generate soliton-based pulses with picosecond-scale durations [33], offering a compact route to MIR pulse generation. However, QCL emission is often limited to wavelengths above $3.9\text{ }\mu\text{m}$, leaving part of the MIR spectral region inaccessible to QCL-based combs. In addition, QCL devices generally exhibit high electrical power consumption and demand stringent thermal management, which further complicates their use in compact sensing platforms. Finally, identifying two QCLs with sufficiently well-matched characteristics for dual-comb generation can be challenging and costly, particularly because fabrication-induced variations can shift their emission wavelengths and limit the availability of suitably matched devices. Other approaches include interband cascade laser frequency combs [34–36]; however, these sources typically still exhibit limited output power and bandwidth, restricted tunability of the comb center wavelength, and comparatively high noise levels.[37] In contrast, nonlinear frequency conversion of near-infrared light in bulk periodically poled lithium niobate (PPLN) crystals can provide broad spectral coverage, but it generally relies on bulky free-space setups and requires high pump powers [38], limiting its suitability for practical and versatile spectroscopic platforms. Some demonstrations have employed an optical parametric oscillator to generate a frequency comb extending up to $3.3\text{ }\mu\text{m}$. But, it requires a high-power pulsed pump source with careful preparation associated with a fixed free spectral range (FSR) matched to that of the ring resonator [39].

More generally, QCL combs rely on complex nonlinear intracavity dynamics, whereas OPO combs are additionally sensitive to cavity detuning, dispersion, and parametric dynamics. These constraints motivate alternative MIR-comb architectures that combine high coherence, broad tunability, and scalable integration. EO combs are particularly well suited to this purpose, as their coherence and comb-line spacing are directly defined by the seed laser and RF drive. They therefore provide deterministic electronic control of the comb spacing and a high degree of reconfigurability.

In this work, we demonstrate efficient MIR electro-optic frequency-comb generation on an integrated thin-film lithium niobate platform. A telecom electro-optic comb is first generated by phase modulation and subsequently down-converted to the MIR through difference-frequency generation in an integrated PPLN section, where it is mixed with a near-infrared pump. We implement this architecture on both LNOI and LNOS, providing a proof of concept for integrated MIR electro-optic comb generation and a direct demonstration of the potential of LNOS for long-wavelength MIR nonlinear photonics due to its larger transparency window. On both LNOI and LNOS, we generate MIR combs with an instantaneous bandwidth of approximately 6 nm

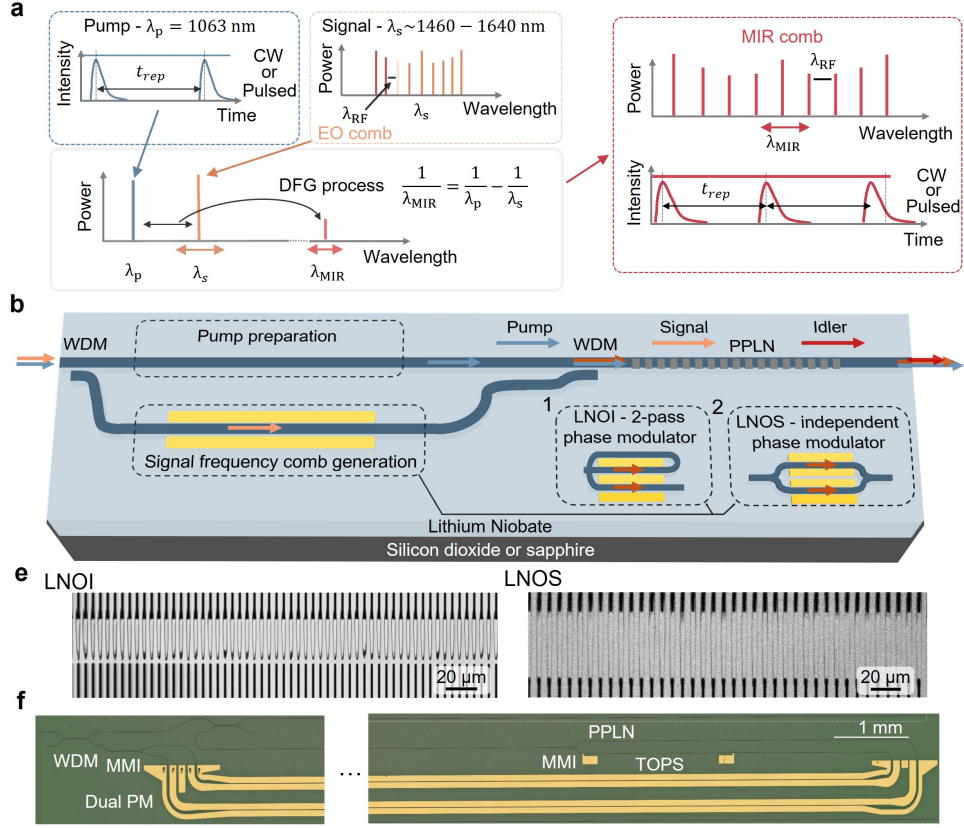


Fig. 1 Working principle of the demonstrated mid-infrared comb generator. **a**, Conceptual diagram of the conversion process. A continuous-wave pump at 1063 nm and a tunable signal around 1550 nm are coupled into a thin-film lithium niobate chip, where a frequency comb near 1550 nm is generated. The pump is mainly operated in continuous-wave (CW) mode but can be modulated either externally or using an integrated amplitude modulator, thereby enabling modulation of the generated MIR output. The pump and signal comb are subsequently combined with an on-chip wavelength-division multiplexer (WDM) and routed to a PPLN section, where difference-frequency generation produces a mid-infrared frequency comb. **b**, Schematic of the fabricated chip, highlighting the functional sections used for pump preparation, signal comb preparation, wavelength combining, and nonlinear conversion in the PPLN section. The inset 1 and 2 shows the telecom signal single and dual frequency comb preparation sections as implemented on LNOI and LNOS, respectively. **c**, Two-photon microscopy images of the poled region on LNOI (top) and LNOS (bottom), revealing the domain inversion. The dark line separation highlights the domain boundaries. **d**, Optical microscope image of a fabricated device on LNOS.

and a free spectral range of up to 12.5 GHz, limited by the radio frequency amplifier. We demonstrate MIR comb generation over wavelength ranges of 3.15–3.7 μm on a single LNOI chip and 3.3–3.9 μm on LNOS. Thermal tuning of the PPLN devices enables phase-matching wavelength shifts of up to 55 nm in LNOI and 120 nm in LNOS. Furthermore, MIR generation at wavelengths up to 4.5 μm is achieved in a

straight LNOS waveguide, highlighting the suitability of this platform for extending integrated lithium-niobate frequency conversion toward longer MIR wavelengths. In this architecture, the comb spacing is set by the RF drive frequency, whereas the comb center frequency is independently tuned through the telecom signal wavelength. This enables independent control of the MIR comb center frequency and free spectral range within a single device. Moreover, on the LNOS platform, we used two independently driven phase modulators to generate an on-chip dual comb. The resulting multiheterodyne beat notes can be detected with a low-bandwidth MIR photodetector across a 150 nm comb center-frequency tuning range, offering a compact alternative to bulky, high-resolution optical spectrometers for MIR spectroscopy. We further demonstrate tunability of the frequency offset between the two combs via serrodyne modulation on one arm. Because both the telecom-band and MIR combs are simultaneously available at the chip output, the platform also enables dual-band spectroscopy for molecules with absorption features in both the NIR and MIR regions.

2 Results

2.1 MIR comb generator presentation

Figures 1a and 1b illustrate the operating principle and schematic of the device, respectively, for both the 0.6 μm film lithium-niobate-on-insulator and 0.9 μm film lithium-niobate-on-sapphire platforms. This work aims to compare the potential of LNOS for efficient MIR generation against that of the more commonly used LNOI platform. The input waveguide top width is 0.8 μm on LNOI and 0.5 μm on LNOS, with corresponding etch depths of 0.4 μm and 0.6 μm , respectively. The waveguides are designed for transverse-electric (TE) single-mode operation at 1550 nm. After being coupled into the chip using a lensed fiber, the 1063 nm continuous wave pump and 1550 nm signal are subsequently split using a wavelength division multiplexer (WDM). On the LNOI platform, the pump is routed through an integrated Mach-Zehnder modulator (MZM), which can be optionally used to switch between operation with a CW or an amplitude-modulated pump. On the LNOS chip, no such modulator was added to the pump path, for the sake of simplicity in device characterization. The signal frequency comb generation is detailed in the insets of Figure 1b. On LNOI, the signal is routed through a double-pass phase modulator to generate a single EO comb (inset 1). On LNOS, the signal is further split by a 1×2 multimode interferometer (MMI) and routed through two independently driven phase modulators, enabling on-chip dual-comb generation (inset 2). A second WDM subsequently recombines the pump and signal fields. The waveguide is then tapered to a top width of 2.5 μm on LNOI and 1.9 μm on LNOS. These widths represent a compromise between minimizing MIR propagation losses associated with mode delocalization and maintaining efficient difference frequency generation (DFG). The taper length is optimized to avoid mode crossings (see Supplementary Material D) and to minimize optical reflections. Finally, the pump and signal are routed through a PPLN section, where the signal EO comb is converted to the MIR through DFG. Figure 1e shows two-photon microscope images of the PPLN region for LNOI (left) and LNOS (right). The poling period in the PPLN region, around 4.9 μm on LNOI (around 7.1 μm on LNOS), is chosen to achieve phase

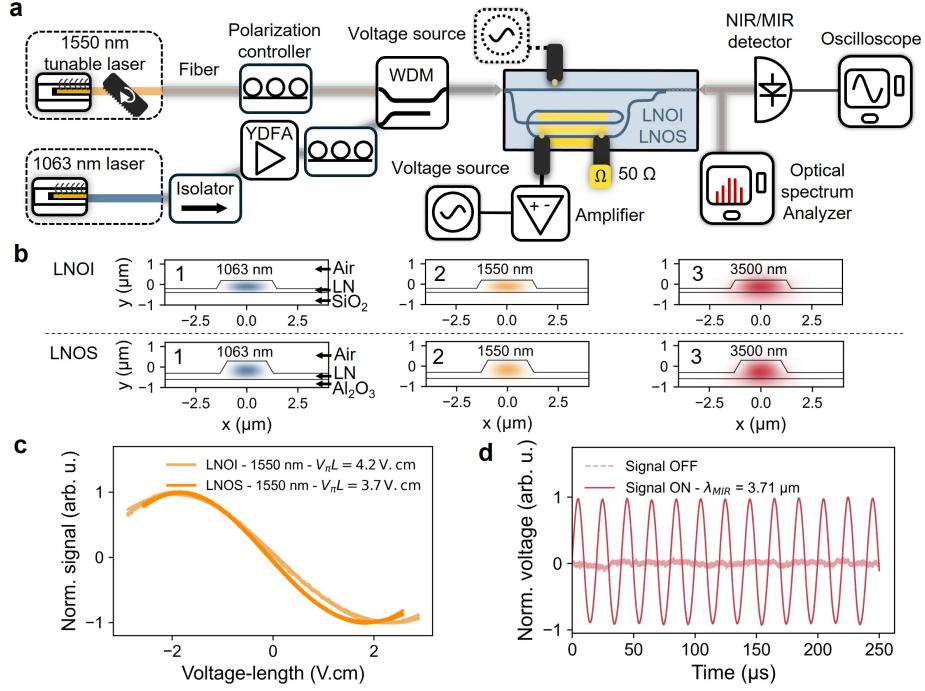


Fig. 2 Characterization setup for the MIR integrated comb generator. **a**, Schematic of the setup used to characterize the fabricated device. The pump laser delivers up to 500 mW at 1063 nm, while a tunable laser provides the 30 mW signal near 1550 nm; additional details on the sources are given in the Methods section. Each laser passes through a polarization controller, with the pump first being amplified by an Ytterbium Doped Fiber Amplifier (YDFA). The two sources are then combined using a fiber-based WDM and coupled into the waveguide through a lensed fiber, with the input polarization adjusted to excite the fundamental TE mode for both the signal and the pump. At the output, the light is collected using an InF₃ multimode lensed fiber. The collected signal is either sent directly to an optical spectrum analyzer (OSA) or a power meter, or alternatively collimated and focused onto a high-speed NIR or MIR photodetector. Two electrical-probe configurations are used for the LNOI and LNOS devices. For single-comb generation on LNOI, a ground-signal-ground (GSG) probe drives the double-pass phase modulator, while a second GSG probe provides a 50 Ω termination to suppress RF reflections. For dual-comb generation on LNOS, a ground-signal-ground-ground-signal-ground (GSGGSG) probe independently drives the two phase modulators. Two single-ended probes are used to bias the thermo-optic phase shifters (TOPS) in the two arms before recombination in the MMI. **c**, The voltage-length product of the 1550 nm modulators on LNOI and LNOS was characterized using a Mach-Zehnder modulator test device. The measured $V_{\pi}L$ values are similar for both platforms, at approximately 4 V cm. **c**, The voltage-length product of the 1550 nm modulators on LNOI and LNOS was characterized using a Mach-Zehnder modulator test device. The measured $V_{\pi}L$ values for both platforms are around the same value at approximately 4 V cm. **d**, MIR amplitude modulation obtained using an externally 50 kHz modulated 1063 nm pump combined with a 1489 nm continuous-wave signal, resulting in modulation of the DFG-generated MIR output.

matching at the desired wavelength. Finally, we demonstrate DFG generation up to 4.2 μm using a straight waveguide with the waveguide geometry as the devices, and

up to 4.5 μm by increasing the waveguide top width (Supplementary Material D). A complete optical micrograph of the LNOS chip is provided in Figure 1f.

2.2 Comb generator performance characterization

Figure 2a shows the experimental setup used to characterize the comb generators. Light is coupled into the waveguide fundamental TE mode using a single-mode lensed fiber. At the output, the light is collected with an InF_3 mid-infrared lensed fiber and routed either to an optical spectrum analyzer (OSA) or a optical power meter. Alternatively, the output light is collimated using a fiber collimator and focused onto a NIR or MIR photodetector with a high-numerical-aperture germanium lens. Figure 2b shows the corresponding optical modes in the PPLN region. Additional simulations of the nonlinear conversion efficiency are provided in Supplementary Material D. As shown in Figure 2c, the modulators fabricated on LNOI and LNOS exhibit $V_\pi L$ values of 4.2 $\text{V} \cdot \text{cm}$ and 4.4 $\text{V} \cdot \text{cm}$, respectively, at 1550 nm. The effective V_π values of the double-pass phase modulator and the dual-phase modulator (single arm) are 3.1 V and 3.28 V, respectively, accounting for the modulator length. The phase-modulation section is intentionally kept simple compared with other resonant-comb architectures [24]. Maximizing the comb bandwidth is not the primary design objective for these devices, as the comb center frequency can be tuned by adjusting the input-signal wavelength. Further characterization of both platforms is provided in Supplementary Materials B and C.

MIR amplitude modulation is demonstrated in Figure 2d, where the 1063 nm pump is externally modulated at 50 kHz via direct AC modulation of the diode current and combined with a 1489 nm continuous-wave signal, resulting in the corresponding modulation of the DFG-generated 3.72 μm MIR output on the LNOS chip. For the LNOI chip, an integrated MZM was implemented on the pump arm to enable modulation of the MIR output. The modulation disappears when the signal is turned off, even though the modulated pump remains present, confirming that the observed signal indeed corresponds to the generated MIR light.

For both the LNOI and LNOS platforms, the generated MIR spectra were measured using an OSA. Figure 3a shows the MIR DFG efficiency as a function of the generated MIR wavelength for multiple devices on each platform, labelled I for LNOI and S for LNOS. The efficiency is normalized by the product of the measured output pump and signal powers, as defined in the Methods section. During the measurement, the CW pump wavelength was fixed at approximately 1063 nm, while the tunable external-cavity laser operating near 1550 nm was swept from 1460 to 1640 nm in 1 nm steps, with an output power of approximately 10 mW.

For LNOI, two PPLN designs are considered. The first is a segmented structure comprising four 0.2 cm-long sections with distinct poling periods, designed to provide broadband phase matching in the mid-infrared (Device I1). This quasi-phase-matching scheme enables broadband DFG, yielding MIR generation over a spectral range of approximately 200 nm, centered at 3.2 μm . The second design consists of a single-period, 0.8 cm-long PPLN section providing phase matching centered at 3.6 μm (Devices I2–I4). Emission around 3.4 μm could be also be obtained by tuning the PPLN period. For these LNOI devices, the longer interaction length combined with the

fixed poling period provides higher nominal nonlinear gain but results in a narrower phase-matching bandwidth. These devices generate MIR light centered near $3.6\ \mu\text{m}$ over a spectral range of approximately $100\ \text{nm}$. However, the conversion efficiency decreases substantially at wavelengths beyond $3.5\ \mu\text{m}$. This reduction is attributed to the increased optical loss associated with the SiO_2 substrate, as indicated by the trend shown in the lower panel of Figure 3a. The losses are further exacerbated by the thinner LN film used in the LNOI platform, which results in weaker mode confinement. Together, these effects offset the gain in conversion efficiency expected from the longer PPLN interaction length.

In order to overcome this limitation of the LNOI platform, we implemented the same single-period design on LNOS, where the sapphire substrate using a $1\ \text{cm}$ -long PPLN section with a poling period optimized for phase matching near $3.6\ \mu\text{m}$ (Devices S1–3). After accounting for the difference in PPLN interaction length, the demonstrated LNOS devices exhibit an approximately $10\ \text{dB}$ higher MIR generation efficiency than the LNOI devices at $3.6\ \mu\text{m}$, increasing to $32.5\ \text{dB}$ at $3.7\ \mu\text{m}$. For Device S1, we obtain a maximum generated MIR power of $2.1\ \mu\text{W}$. Temperature tuning provides an additional degree of control over the phase-matching condition and extends the accessible MIR wavelength range of any device. The phase-matched idler wavelength shifts by approximately $0.5\ \text{nm}/^\circ\text{C}$ on LNOI and $1.2\ \text{nm}/^\circ\text{C}$ on LNOS as shown in the Figure 3b. The difference is attributed to sapphire having a larger thermo-optic coefficient than SiO_2 . Within the maximum operating temperature of $160\ ^\circ\text{C}$ imposed by our experimental setup, the MIR emission can be red-shifted by up to $70\ \text{nm}$ on LNOI and $168\ \text{nm}$ on LNOS.

2.3 MIR electro-optic comb generation and nonlinear conversion.

For MIR comb generation, the phase modulators were driven by an RF signal with an input power of $40\ \text{dBm}$. A representative MIR EO comb spectrum generated on LNOI is shown in Figure 3c. The comb is centered at $3.287\ \mu\text{m}$ and comprises 12 sidebands spaced by $12.5\ \text{GHz}$. It spans an instantaneous bandwidth of approximately $6\ \text{nm}$, with a measured power per comb line of approximately $-52\ \text{dBm}$. Figure 3d shows an MIR EO comb generated on LNOS, centered at $3.715\ \mu\text{m}$. In this case, the power contrast between the central carrier and the modulation sidebands is substantially larger than in the LNOI device. This behavior is attributed to an imbalanced WDM, which directs only a fraction of the telecom signal power into the phase-modulation arm, as discussed in Supplementary Material C. This limitation is not intrinsic to the LNOS platform and can be easily addressed through an optimized WDM design with a higher splitting ratio at the telecom wavelength.

2.4 MIR dual-comb EO comb generation

The implemented LNOS architecture enables on-chip dual-comb generation by using two independently driven phase modulators to produce electro-optic combs with slightly different free spectral ranges Δ_{MW} . As illustrated in Figure 4a, two amplified

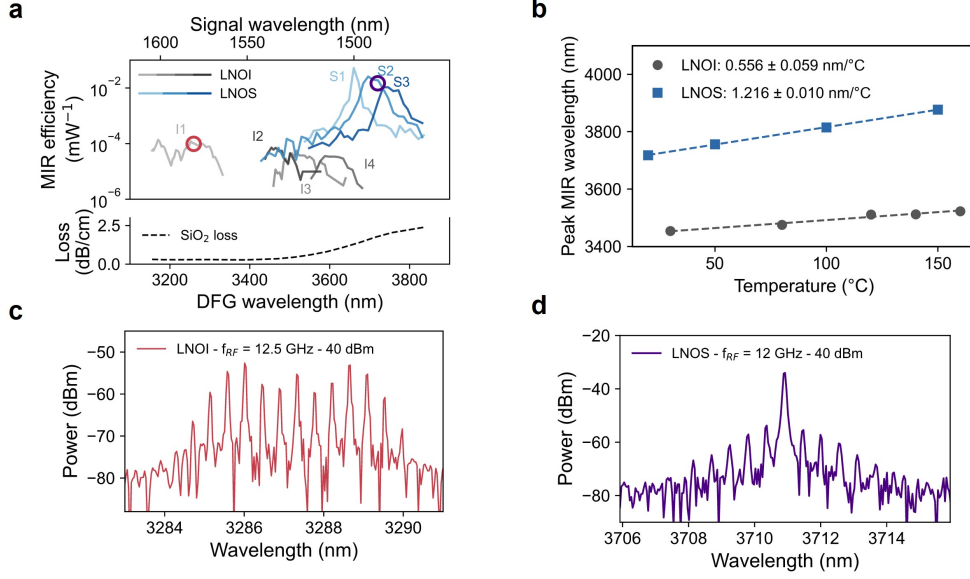


Fig. 3 Experimental demonstration of MIR comb generation with broadband difference-frequency generation. **a**, Illustration of the MIR wavelength coverage provided by the two device platforms LNOI and LNOS, spanning approximately 3.15-3.9 μ m. The graph shows the MIR power normalized to the transmitted pump and signal at the chip output. The bottom plot also shows material absorption of SiO₂ as a function of wavelength. **b**, Phase matching of the PPLN as tuned by temperature for LNOI and LNOS. The thermal tuning is shown to be roughly twice as strong on LNOS as compared to the LNOI. **c**, Example of a MIR electro-optic frequency comb generated on LNOI with 40 dBm RF drive power, centered around 3.287 μ m at a modulation frequency of 12.5 GHz. **d**, Example of a MIR electro-optic frequency comb generated on LNOS with 40 dBm RF drive power, centered around 3.711 μ m at a modulation frequency of 12 GHz.

and phase-locked RF sources independently drive the two telecom phase modulators through a GSGSG probe. To provide fine control over the relative carrier offset between the two combs, we implemented serrodyne modulation by mixing a low-frequency signal with the RF drive applied to one of the phase modulators. This produces a continuous linear phase ramp, which introduces a relative difference between the carrier frequency of the two combs. On-chip, the two NIR combs are subsequently recombined with an MMI and translated to the MIR through DFG with the 1063 nm pump, as shown in Figure 4b. The resulting MIR dual comb is detected using a low-bandwidth (1.5 MHz) InAsSb photodetector, with a germanium filter ensuring that only the MIR comb is detected. Interference between corresponding teeth of the two combs produces a multiheterodyne signal at the difference between their free spectral ranges, Δ_{MW} , thereby down-converting the optical comb structure to radio frequencies. Several verification tests were performed to confirm that the detected signal originated solely from the MIR dual comb and not from the telecom-generated dual comb: turning off the pump source, or tuning the telecom signal away from the phase-matching condition, caused the heterodyne beat note to disappear, as expected.

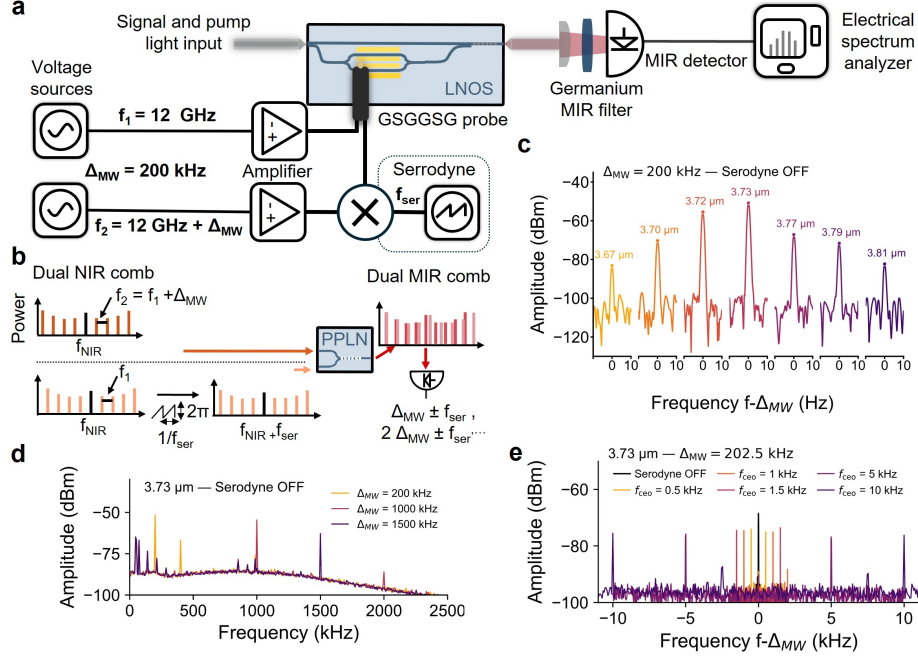


Fig. 4 Experimental demonstration of dual-comb MIR generation. **a**, Experimental setup for on-chip dual-comb generation. Two amplified, phase-locked RF sources independently drive the two phase modulators through a GSGGSG probe. A mixer injects serrodyne modulation into one of the phase modulators, enabling tunable control of the frequency offset between the two combs. **b**, On-chip recombination and MIR generation. The two NIR combs are combined with an MMI and translated to the MIR through DFG with the 1063 nm pump. **c**, ESA measurements of the multi-heterodyne beat notes produced by dual-comb electro-optic modulation with repetition-rate differences of 200, 1000, and 1500 kHz for a generated MIR signal around 3.73 μm . In this case, serrodyne modulation is off. **d**, ESA measurements of the 200 kHz beat note for different MIR comb center wavelengths using Device S3. The beat note is observed over a comb center-wavelength tuning range of 140 nm and exhibits a linewidth below 5 Hz, demonstrating the high spectral purity of the generated MIR dual comb. In this case, serrodyne modulation is off. **e**, ESA measurements of the beat note as the frequency of the applied serrodyne signal is tuned from 0.5 kHz to 10 kHz, demonstrating fine control over the comb offset frequency.

Figure 4c shows the electrical spectra obtained for a comb spacing differences of 200, 1000, and 1500 kHz. In each case, the fundamental multi-heterodyne beat note and its second harmonic are clearly resolved, confirming the generation and MIR detection of the dual-comb signal. Figure 4d shows the 200 kHz beat note measured for different MIR comb center wavelengths using Device S3. The beat note remains detectable over a 140 nm tuning range of the comb center wavelength and exhibits a linewidth below 5 Hz. The accessible spectral range could be further extended through thermal tuning of the phase-matching condition. This narrow linewidth demonstrates the high spectral purity of the generated MIR dual comb. Finally, Figure 4e shows that

serrodyne modulation enables electronic tuning of the relative carrier-frequency offset between the two combs from 0.5 kHz to 10 kHz. Without serrodyne modulation, beat notes arising from comb-tooth pairs with positive and negative relative detunings overlap at the same beat-note frequency, Δ_{MW} , when detected on a photodetector. Consequently, the individual contributions to the measured beat note cannot be distinguished. Applying serrodyne modulation to one comb shifts all of its teeth by a controlled frequency f_{ser} . The previously overlapping contributions are thereby separated into two resolved beat notes at $\Delta_{\text{MW}} \pm f_{\text{ser}}$, enabling unambiguous identification of positive and negative comb-tooth detunings. This small, electronically tunable offset removes the ambiguity without requiring moving parts or thermal actuation, while preserving compatibility with low-bandwidth photodetection through the ability to set the offset to a controlled small value.

3 Discussion

Our integrated electro-optic comb generator provides a flexible route to MIR frequency-comb generation, with independent control of the comb center frequency, the frequency offset and line spacing within a single device. The comb center wavelength can be efficiently tuned over several hundred nanometres by adjusting the telecom seed wavelength and the quasi-phase-matching condition, while the free spectral range is set electronically by the RF modulation frequency. Thermal tuning of the chip allows the phase-matching window to be shifted by up to approximately 150 nm. In contrast to many MIR comb-generation approaches based on high-power pulsed pump sources, our architecture operates with two continuous-wave diode lasers, delivering 27 dBm at 1063 nm and 10 dBm at 1550 nm. This approach benefits from the low power consumption, compactness, and technological maturity of near-infrared and telecom-band laser sources. The spectral coherence of the MIR comb is inherited from the seed lasers and RF drive signals. The direct comparison between LNOI and LNOS further demonstrates the advantages of the sapphire substrate for long-wavelength MIR nonlinear conversion. In particular, LNOS provides improved nonlinear conversion efficiency beyond 3.5 μm , where the SiO_2 buried oxide layer in LNOI introduces increasing optical losses. The LNOS platform therefore enables extended MIR wavelength coverage while maintaining efficient DFG. Using straight waveguides, we demonstrate MIR generation at wavelengths up to 4.5 μm . The architecture enables on-chip MIR dual-comb generation using two independently driven phase modulators. The MIR comb center wavelength can be tuned over 140 nm while maintaining a detectable multiheterodyne beat note. Serrodyne modulation applied to one of the phase modulators further provides a tunable frequency offset between the two combs, resolving the sign ambiguity of the beat note and allowing its frequency to be positioned at very low frequencies within the bandwidth of a low-bandwidth detector. Within the same integrated platform, the device combines broad spectral accessibility with precise electronic control of the comb spacing through the RF drive frequency and of the relative carrier-frequency offset between the two combs. This is particularly advantageous for spectroscopy, where molecular absorption features may occupy relatively narrow spectral windows: the comb center frequency can be tuned to place the available power

per comb tooth within the spectral region of interest. Furthermore, on-chip dual-comb generation down-converts the optical spectrum to low-frequency multiheterodyne beat notes, removing the need for a bulky, high-resolution MIR optical spectrum analyzer. Instead, the spectrum can be measured using a low-bandwidth MIR photodetector and an electrical spectrum analyzer. The simultaneous availability of NIR and MIR combs at the chip output also makes the platform attractive for multi-wavelength sensing, enabling complementary detection of molecules with absorption features in both spectral regions. For example, methane exhibits widely used absorption bands near 1.65 μm and 3.3 μm . Further improvements in conversion efficiency could be achieved through adaptive poling, which locally optimizes the quasi-phase-matching condition to enhance the generated idler power, generally at the expense of reduced spectral bandwidth. Expanding the comb bandwidth would also enable broadband dual-comb spectroscopy without requiring extensive tuning of the signal wavelength. Future work will focus on applying this platform to dual-comb spectroscopy measurements. These results establish a scalable pathway towards broadband, tunable, and practically deployable on-chip MIR comb sources for spectroscopy.

4 Methods

4.1 Device fabrication

The devices are fabricated from commercially available x-cut lithium-niobate-on-insulator (LNOI, NanoLN) and lithium-niobate-on-sapphire (LNOS, NGK Insulators) wafers. The LNOI wafers comprise a 0.6 μm -thick lithium niobate film on a 2 μm -thick buried SiO_2 layer, whereas the LNOS wafers comprise a 0.9 μm -thick lithium niobate film on a sapphire substrate. Periodic poling is first performed on the unetched lithium niobate film. Chromium poling electrodes with the desired period are defined by electron-beam lithography and metal lift-off. High-voltage pulses are subsequently applied to selectively invert the ferroelectric domains to form the PPLN region. This periodic domain inversion modulates the sign of the nonlinear coefficient which can be used to engineer quasi-phase matching [40]. The domain inversion is verified by two-photon microscopy, after which the poling electrodes are removed via wet etching. For waveguide fabrication on LNOI, waveguides are first patterned in hydrogen silsesquioxane (HSQ) resist via electron-beam lithography. The waveguides are then etched by argon-based inductively coupled plasma reactive-ion etching (ICP-RIE). The waveguide top width increases from 0.8 μm in the light-preparation section to 2.5 μm in the frequency-conversion section, with an etch depth of 0.4 μm . The etch redeposition is then removed using a potassium hydroxide (KOH) cleaning step, followed by a buffered oxide etch to remove the residual HSQ mask. For waveguide fabrication on LNOS, waveguides are first patterned in a polymer resist via electron-beam lithography. The waveguides are then etched by argon-based ICP-RIE. The waveguide top width increases from 0.5 μm in the light-preparation section to 1.9 μm in the frequency-conversion section, with an etch depth of 0.6 μm . The etch redeposition is then removed using an RCA-SC1 cleaning step, followed by cleaning with Piranha solution and a buffered oxide etch to remove the residual mask. On both substrates, the anisotropic physical etch produces sidewall angles of approximately

60°. To reduce etch-induced damage and optical propagation losses, the devices are annealed at 500 °C under ambient conditions for 2 h. Thermo-optic phase shifters are fabricated by electron-beam lithography using a PMMA/MMA bilayer, followed by the evaporation of a 100 nm-thick gold layer (Evatec 50) and lift-off. The electro-optic electrodes are subsequently patterned by direct laser writing (MLA 150), followed by the evaporation of a 900 nm-thick gold layer and lift-off. The electrodes are designed to provide a microwave impedance of 50 Ω for high-speed modulation. Finally, the chips are diced using a Disco DAD 3221 to expose the waveguide facets. The facets are mechanically polished using diamond abrasive (Allied Multiprep System 8) to reduce input and output coupling losses and are subsequently refined by focused-ion-beam milling (TFS Helios 5 UX) to improve their quality.

4.2 Design of the device

The passive photonic components, namely the waveguides, MMIs, crossing and grating coupler for characterization, were first simulated with an eigenmode expansion and finite element solver (Ansys Lumerical) to determine suitable geometries. Electro-optic performance was then evaluated with COMSOL Multiphysics and Ansys Lumerical, focusing on modulation efficiency, RF-optical field overlap, and thermo-optic behavior. Starting from the waveguide geometry optimized for single-mode guidance and a chosen gap, the widths and spacings of the transmission lines were refined through RF-optical co-simulations in COMSOL Multiphysics to achieve 50 Ω impedance and velocity matching with the optical mode, thereby suppressing reflections and enhancing the modulation bandwidth. Previous characterization has confirmed that the presented radio-frequency design demonstrates bandwidths exceeding 20 GHz.[28]

4.3 Electro-optic characterization

The pump source is a distributed Bragg reflector (DBR) laser diode emitting near 1063 nm, with a maximum output power of 10 mW (Thorlabs DBR1064P) and a linewidth of approximately 5 MHz. Its output is subsequently amplified by a polarization-maintaining YDFA with a saturation power of 26 dBm (Civil Laser YDFA-26-PM-M). The signal source is a TOPTICA DLC tunable diode laser delivering up to 30 mW over the 1460–1560 nm wavelength range. Its intrinsic linewidth is on the order of 100 Hz, although its observed linewidth is approximately 5 MHz due to frequency drift. In specific cases, a Keysight Newport laser (N7776C) was used to access longer signal wavelengths from 1490 to 1640 nm, providing an output power of around 10 mW and a linewidth of approximately 50 MHz. Optionally, the signal can be amplified using an Erbium Doped Fiber Amplifier (EDFA) with a saturation power of 33 dBm but only power up to 25 dBm have been used. To evaluate the modulation efficiency, static measurements were performed on a standalone test MZM using grating-coupler coupling. The optical path difference between the two arms is thermally tunable via the integrated TOPS, which was characterized by sweeping the electrical power and monitoring the resulting change in the MZM output power. The half-wave voltages V_π at 1550 nm were extracted by driving the modulator with a triangular waveform 1 kHz. The modulated optical signal was detected with an InGaAs

photodetector (New Focus 2053), recorded on a digital oscilloscope, and analyzed using a custom Python routine.

4.4 MIR light generation using difference frequency generation

To measure generated MIR light at different center wavelengths, the signal-laser wavelength was swept and the corresponding output spectra were recorded using an OSA. The modulated output was collected with a MIR lensed fiber and routed to the OSA for spectral analysis. To compare measurements acquired at different pump and signal powers on chip, which may vary due to facet quality and device-to-device variations, we normalized the measured MIR power to the total output power at 1063 nm and at the considered NIR wavelength λ_{NIR} . The normalized MIR efficiency is defined as

$$\eta_{\text{MIR}}(\lambda_{\text{MIR}}) = \frac{P_{\text{MIR}}(\lambda_{\text{MIR}})}{P_{1064} P_{\text{NIR}}(\lambda_{\text{NIR}})}. \quad (1)$$

Here, $P_{\text{MIR}}(\lambda_{\text{MIR}})$ is the measured MIR output power at the DFG wavelength λ_{MIR} , expressed in mW. The corresponding NIR signal wavelength is denoted by λ_{NIR} . The quantities P_{1064} and $P_{\text{NIR}}(\lambda_{\text{NIR}})$ are the measured output powers, expressed in mW, at 1064 nm and at λ_{NIR} , respectively. Consequently, the normalized MIR efficiency $\eta_{\text{MIR}}(\lambda_{\text{MIR}})$ is expressed in mW^{-1} .

4.5 MIR electro-optic comb generation and characterization

For efficient single comb signal generation on the LNOI platform, we implemented a double-pass phase-modulation scheme, as illustrated in Figure 1b. This configuration effectively increases the modulation length without requiring a higher RF drive power. The RF signals were generated externally, amplified using a high-power RF amplifier (RF-Lambda), and delivered to the on-chip travelling-wave electrodes using a high-speed GSG probe. The electrode termination was matched to 50Ω to suppress RF reflections. For both platforms, the electrode geometry was chosen to maximize the electro-optic interaction while minimizing plasmonic absorption losses. Maximizing transmitted optical power at 1550 nm is particularly important for efficient subsequent DFG. The electrode width and spacing were further optimized to provide a characteristic impedance of 50Ω and to match the effective indices of the optical and microwave modes. Since the comb bandwidth scales with the phase-modulation index, the double-pass configuration increases the number of generated sidebands at a given RF drive power. On LNOS, two independently driven phase modulators, addressed using a GSGSG probe and incorporating electrodes approximately 1 cm long, are employed to generate the two NIR combs required for dual-comb operation. The same RF-design considerations were applied to this modulator.

Supplementary information. Further experimental data and analysis can be found in the Supplementary Information.

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Data availability. The data supporting the plots within this article are available from the corresponding author upon request.

Appendix A Platform fabrication process

Supplementary Figure A1 presents a schematic of the fabrication flow, described in detail in the Methods section, used to realize thin-film lithium niobate photonic circuits incorporating a PPLN section. The inset of the Figure A1 shows a top view of the periodically poled region, with white arrows indicating the orientation of the ferroelectric domains.

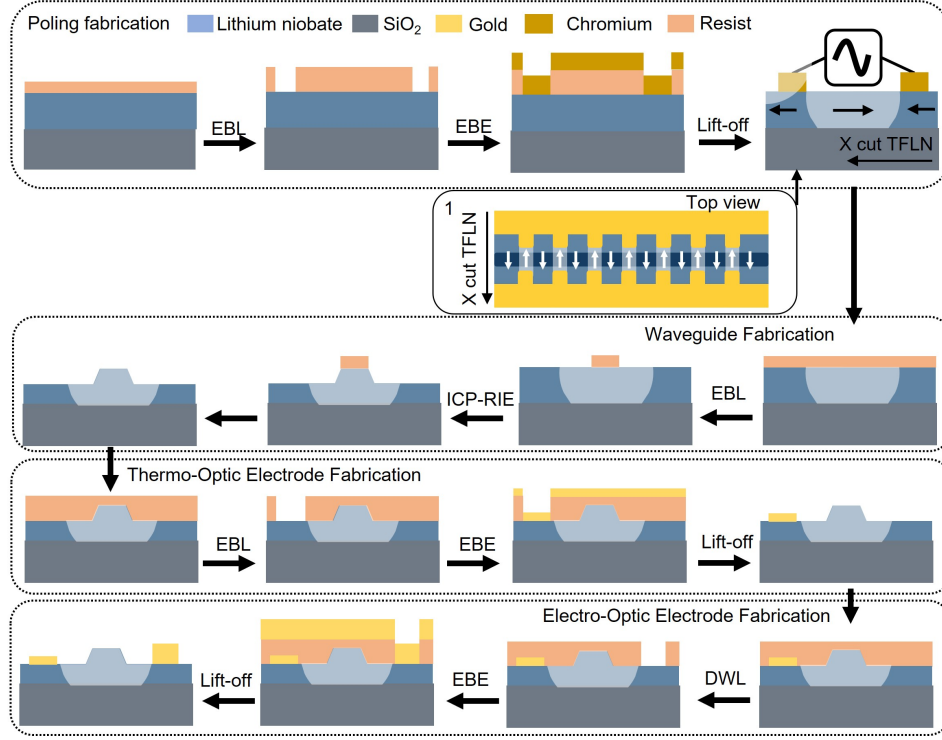


Fig. A1 Fabrication process of the TFLN photonic circuits Fabrication process for lithium niobate on insulator with PPLN section. EBL: electron-beam lithography; ICP: inductively coupled plasma; EBE: electron-beam evaporation; DWL: direct-write laser lithography. For the poling fabrication section, the finger-shaped electrodes are deposited on the LN thin film, and high-voltage pulses (approximately 1,kV) are applied to invert the ferroelectric domains, thereby forming the periodically poled region as shown in Inset 1. Upon complete poling, domain inversion extends through the full thickness of the LN film.

Appendix B Additional LNOI platform characterization

An optical micrograph of the devices fabricated on LNOI is shown in Figure B2a. The estimated fiber-to-fiber insertion losses are approximately 14 dB at 1063 nm and 15 dB at 1550 nm. These losses are dominated by input- and output-coupling losses and plasmonic absorption in the phase modulator. For component characterization, light at 1063 nm and 1550 nm is coupled on and off the chip using optimized linear grating couplers, ensuring stable coupling conditions and improved measurement reproducibility. The MMI performance is evaluated with four-stage cascaded MMI structures terminated by grating couplers at 1063 nm; the corresponding results are shown in Supplementary Figure B2b. From a linear fit of the transmitted power, the excess loss of the MMIs is extracted to be 0.7 dB at 1063 nm. The grating couplers are

first designed using a commercial FDTD solver, where parameter sweeps over grating period, duty cycle, and emission angle are performed to identify optimal configurations for the target wavelength and polarization. The crossing device loss is characterized using four-stage cascaded crossings placed after a 1×2 MMI splitter, which allows us to remove the contribution of the grating couplers and use a reference arm with identical couplers at 1550 nm; the results in Supplementary Figure B2c indicate that the crossing loss remains below 2 dB over the full wavelength range, and below 1 dB between 1500 and 1600 nm. An important component of the platform is the on-chip 1063/1550 nm DC WDM. As shown in Figure B2d, its performance was characterized using dedicated test structures and grating couplers. The interaction length of the directional coupler was chosen to be 60 μm . By design, 90 % of light at 1063 nm remains in the bar waveguide while 99 % of light at 1550 nm is coupled to the cross waveguide, enabling efficient splitting of the two wavelengths. Finally, the bar-port power was measured from 1490 nm to 1640 nm for a 2×2 directional coupler, as reported in Supplementary Figure B2e; the very low bar-port transmission confirms the good performance of the wavelength-division multiplexing scheme for the 1063 nm and 1550 nm signals.

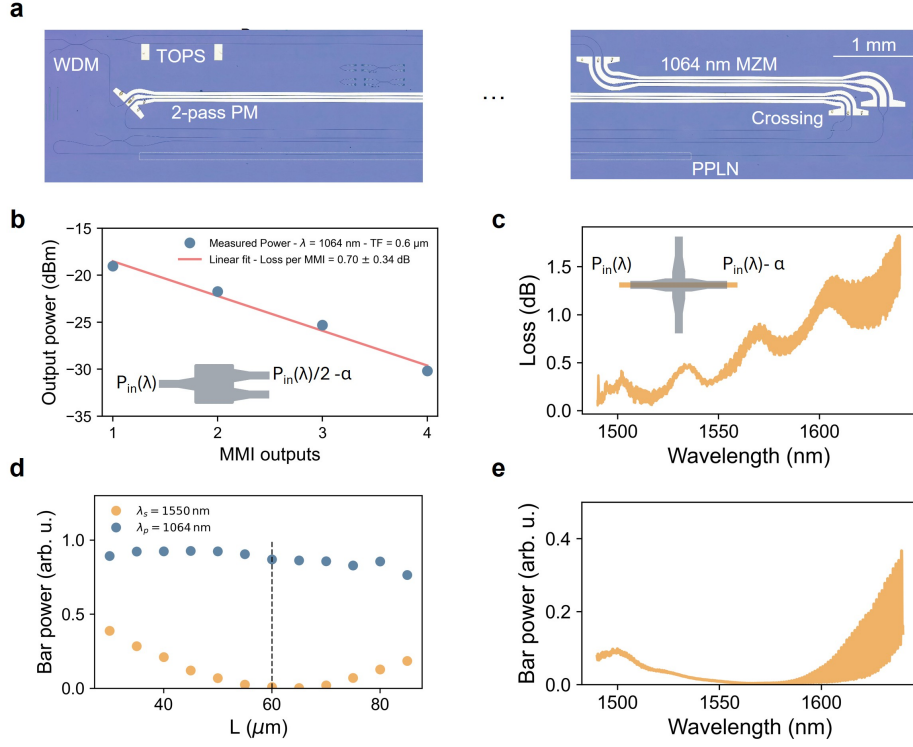


Fig. B2 Characterization of passive components. **a**, Optical microscope image of a fabricated device on LNOI. **b**, Transmission of four-stage cascaded MMIs at 1063 nm, used to extract an excess MMI loss of 0.7 dB. **c**, Crossing-loss measurement using four-stage cascaded crossings with a 1×2 MMI reference arm, demonstrating losses below 2 dB over the full wavelength range and below 1 dB between 1500 and 1600 nm. **d**, Bar port transmitted power for 1063 nm pump and 1550 nm signal as a function of coupling length. **e**, Bar-port power at 1550 nm for a 2×2 directional coupler, indicating negligible bar-port transmission and efficient wavelength-division multiplexing between 1063 nm and 1550 nm.

Appendix C Additional LNOS platform characterization

The estimated fiber-to-fiber insertion losses are approximately 17 dB at 1063 nm and 20 dB at 1550 nm. These losses are dominated by input- and output-coupling losses and plasmonic absorption in the phase modulator. For the LNOS device, additional losses arise from the facet quality and the smaller waveguide top width. For component characterization, light at 1063 nm and 1550 nm is coupled onto and off the chip using optimized linear grating couplers, ensuring stable coupling conditions and improved measurement reproducibility. The performance of the MMIs used for dual-comb recombination on LNOS is evaluated using four-stage cascaded MMI structures terminated by grating couplers at 1063 nm; the corresponding results are shown in Supplementary

Figure C3a. From a linear fit of the transmitted power, the excess loss of the MMIs is extracted to be 0.3 dB at 1550 nm. The grating couplers were first designed using a commercial FDTD solver, in which parameter sweeps over grating period, duty cycle, and emission angle were performed to identify optimal configurations for the target wavelength and polarization. As shown in Figure C3b, we characterize the Q factor of several 150 μm ring resonators with varying gaps to reach critical coupling, in order to evaluate the propagation loss at the telecom wavelength on the LNOS platform. We obtain a Q factor of approximately 290,000, corresponding to a FSR of 1.17 nm and an extinction ratio around 20 dB. This allows the propagation loss to be extracted over the 1490–1580 nm range, where it remains below 0.7 dB/cm, with the lowest recorded loss of approximately 0.5 dB/cm near 1500 nm, as shown in Figure C3c. An important component of the platform is the on-chip 1063/1550 nm WDM. As shown in Figure B2d, its performance was characterized using dedicated test structures and grating couplers. The interaction length of the directional coupler was swept across the different devices on the LNOS chip, ranging from 50 μm to 1000 μm . By design, the 1063 nm light remains predominantly in the input (bar) waveguide with an efficiency of approximately 90%, whereas the 1550 nm light does not fully couple to the cross waveguide at any of the tested interaction lengths. This incomplete coupling is currently unexplained and is likely responsible for the discrepancy between the main carrier peak and the comb line power, since a non-negligible fraction of the 1550 nm power remains unmodulated.

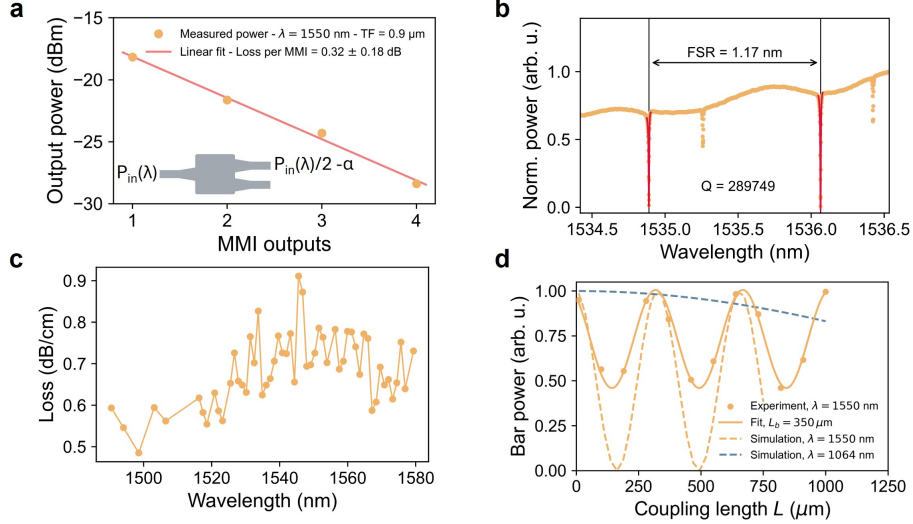


Fig. C3 Characterization of passive components for the LNOS platform. **a**, Transmission of four-stage cascaded MMIs at 1063 nm, used to extract an excess MMI loss of 0.7 dB. **b**, Measured ring resonator spectra around 1535 nm, showing a Q-factor of around 290,000 and an FSR of 1.17 nm. **c**, Calculated waveguide propagation losses in the telecom C-band from ring resonator spectra, showing losses below 1 dB/cm across the full range. **d**, Bar-port power at 1550 nm for a 2x2 directional coupler, indicating negligible bar-port transmission but only partial coupling of the 1550 nm light, hence limiting the WDM efficiency.

Appendix D Nonlinear PPLN simulation

Figure D4 presents additional simulation for the PPLN region design. Figure D4a shows the poling period required to satisfy the quasi-phase-matching condition as a function of idler wavelength and waveguide top width on LNOI. Figure D4b presents the corresponding normalized conversion efficiency (CE), revealing a mode-crossing region where the efficiency drops sharply. Below this mode-crossing region, the mid-infrared mode is weakly confined and highly lossy; we therefore selected a waveguide width above the mode crossing, maintaining a safety margin to account for potential deviations from the simulations or variation in LN film thickness. Equivalent results for the LNOS chip are shown in Figure D4c and Figure D4d. The SEM image in Figure D4e reveals a corrugation resulting from the crystal-orientation-dependent etching rate, confirming that the ferroelectric domains extend throughout the entire height of the ridge. Using straight waveguides, we demonstrate MIR generation at wavelengths up to 4.5 μ m D4f. A 1120 nm diode laser was used to access this extended wavelength range. Waveguides with the same top width as the devices presented above generate MIR light up to 4.2 μ m, whereas increasing the top width to $w = 4$ μ m extends the DFG wavelength to 4.5 μ m.

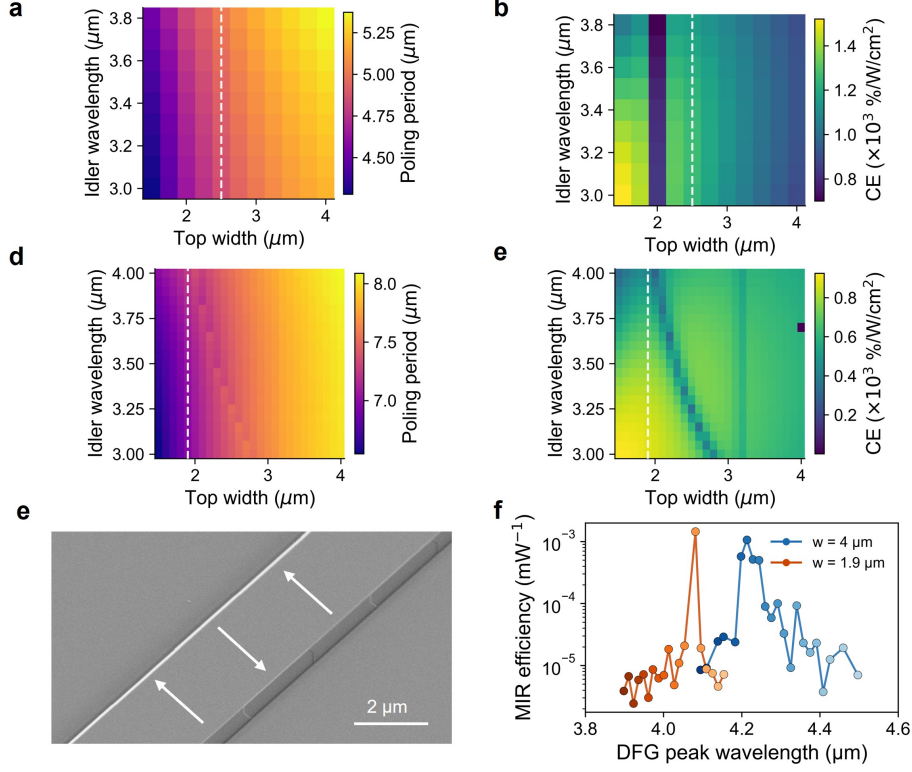


Fig. D4 Simulated dependence of the PPLN waveguide design on top width and idler wavelength for the two platforms. **a**, Required poling period to achieve quasi-phase-matching as a function of idler wavelength and waveguide top width for LNOI chip. **b**, Corresponding normalized conversion efficiency (CE), highlighting a mode-crossing region with a pronounced efficiency drop for LNOI chip. To avoid weak confinement and high loss of the mid-infrared mode below this region, a waveguide width above the mode crossing is selected, with an additional safety margin to account for fabrication and modeling variations. **c**, Required poling period to achieve quasi-phase-matching as a function of idler wavelength and waveguide top width for LNOS chip. **d**, Corresponding normalized CE, highlighting a mode-crossing region with a pronounced efficiency drop for LNOS chip. **e**, SEM image showing the corrugation caused by the dependence of the etching process on the crystal orientation, confirming that the ridge is poled over its entire height. **f**, MIR conversion efficiency as a function of the DFG peak wavelength. For this characterization, a 1120 μm diode laser was used to reach longer wavelengths. The red curve shows the results obtained with a waveguide having the same top width as the devices presented in the main text, while the blue curve shows the results obtained with a wider waveguide ($w = 4 \mu\text{m}$).

Appendix E MIR Dual-tone EO comb generation

Our implementation on the LNOI platform supports the generation of dual-tone combs with two distinct FSRs by driving the phase modulator with two different RF frequencies. In this case the signal has been amplified up to 25 dBm output power to obtain more MIR DFG power. By resolving the heterodyne beat notes on the detector, the

absorption spectrum can be reconstructed. As shown in Figure D4a, two phase-locked RF voltage sources were used. Their outputs were combined using an RF splitter/combiner and subsequently amplified with a single RF amplifier. The resulting RF signal was applied to the phase modulator, and the generated MIR dual-comb spectrum was observed on the optical spectrum analyzer. As seen in Figure D4b, we demonstrate the generation of a dual-tone comb with sideband spacings of 8 GHz and 12 GHz. These spacings were chosen to be well resolved by the optical spectrum analyzer, which has a resolution of approximately 1.9 GHz. To confirm the presence of the dual-tone comb state, we measured the corresponding beat notes on a NIR photodiode. In this case, the difference between the two comb spacings was chosen to be approximately 4 MHz, so as to remain within the limited photodiode bandwidth of about 20 MHz. As highlighted in Figure D4c, the beat notes are clearly resolved. A sufficiently sensitive low-bandwidth MIR photodiode was not available for these measurements.

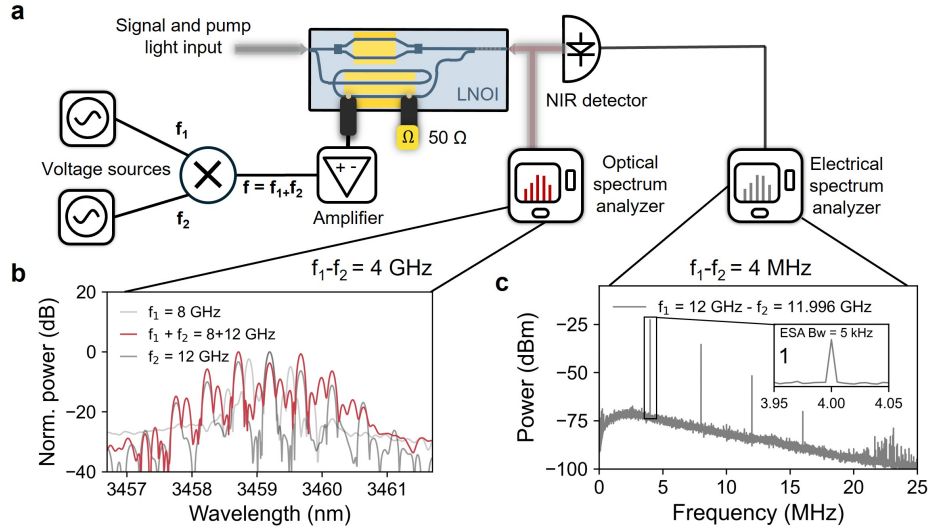


Fig. E5 Experimental demonstration of dual-tone comb generation on the LNOI platform. **a**, Experimental setup used for dual-tone comb generation. Two phase-locked voltage sources are combined using an RF combiner and subsequently amplified to drive a double-pass phase modulator via a GSG probe. **b**, Optical spectrum analyzer measurement showing the first- and second-order sidebands corresponding to electro-optic modulation at 8 GHz and 12 GHz. The modulation frequencies are chosen such that the comb lines are resolvable with the 1.9 GHz resolution of the OSA. **c**, Electrical spectrum analyzer measurement of the beat note between two signals spaced by 4 MHz, corresponding to modulation frequencies of 12 GHz and 11.996 GHz.

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