Efficient frequency comb generation in AlGaAs-on-insulator: supplementary material

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SECTION I. Fabrication of AlGaAs-on-insulator devices

Fig. S1 shows the fabrication process of the AlGaAs-on-insulator (AlGaAsOI) microresonators. Important processes for high Q microresonators include epitaxial material growth, substrate removal, electron-beam lithography (EBL), and dry etching as the waveguide surface smoothness is highly sensitive to those processes.

Fig. S1. Fabrication processes of AlGaAsOI microresonator.

A critical element in the substrate removal process is the choice of etch-stop layer as e.g. hydrofluoric acid etching of Al(Ga)As leaves a too rough surface. Fig. S2 shows the scanning electron microscopy

(SEM) pictures of etched patterns on AlGaAsOI wafers prepared by using different substrate removal processes. Large roughness is observed at the top and bottom surfaces of the AlGaAs film when a conventional AlAs etch-stop layer is used [1] (see Fig. S2(a)). Significant improvement concerning surface smoothness can be observed when a proper etch-stop layer (InGaP in this case) was used as shown in Fig. S2(b).

Fig. S2. SEM pictures of etched patterns on AlGaAsOI wafers fabricated by using different etch-stop layers: (a) AlAs (b) InGaP. AlGaAs material is denoted by the artificial blue colour in both pictures. Etch mask hydrogen silsesquioxane (HSQ) is left on top of AlGaAs patterns. The same dry etching processes are used in both cases.

EBL is used to define the pattern in the HSQ. However, standard EBL process results in severe stitching (denoted by arrows in Fig. S3(a)) between fractured pattern segments, which gives rise to extra roughness on bending patterns. The stitching effect may be due to resist charging in EBL process [2]. To mitigate the stitching effect, multi-pass EBL process was applied and smooth curved waveguide patterning can be realized as shown in Fig. S3(b). The masking HSQ is kept on top of the AlGaAsOI devices since the dielectric properties of HSQ is very similar to SiO₂, which will not adversely affect device performances [3,4]. In addition, the high dose electron-beam-exposed HSQ experiences very similar redistribution of chemical bond as a high temperature annealing

process [5], which convert HSQ to a material with the same index of refraction and chromatic dispersion as SiO_2 [4]. Therefore, we only use SiO_2 as the overcladding material in device dispersion calculation.

Fig. S3. EBL patterning comparison. Top-view SEM pictures of curved pattern in developed e-beam resist HSQ with different EBL processes: (a) standard process, (b) multi-pass process.

SECTION II Characterization of AlGaAs-on-insulator nano-waveguide

Fig. S4(a) shows the SEM cross-section picture of a fabricated AlGaAsOI nano-waveguide cladded in glass. Benzocyclobutene (BCB) was used as bonding layer as denoted by the yellow colour. A strong light confinement can be achieved due to a small effective mode area [6] ($0.16 \mu\text{m}^2$) of the waveguide with cross-section dimension of $320 \text{ nm} \times 630 \text{ nm}$. At the actual device sample facet, the waveguide is tapered from 450 nm to 120 nm as shown in Fig. 4S(b) for efficient fiber-to-chip coupling. We used the modified cut-back measurement to extract propagation loss. Fig. S4(c) shows the measured insertion loss of AlGaAsOI nano-waveguides with different lengths showing a linear loss of 1.4 dB/cm and a fiber-to-chip coupling loss of 2.7 dB/facet . We measured three waveguides for each length and the waveguides used in the measurement have the same number of bends.

Fig. S4. SEM pictures of (a) AlGaAsOI nano-waveguide (denoted in artificial blue colour) cladded in SiO_2 , and (b) nanotaper tip end at the sample facet. (c) Measured insertion loss of AlGaAsOI waveguides with different lengths. (d) Measured four-wave mixing (FWM) conversion efficiency in a 3-mm-long waveguide as a function of pump power.

A four-wave mixing (FWM) measurement was performed using two continuous wave (CW) signals (one for the pump and one for the signal) in a 3-mm-long AlGaAsOI nano-waveguide with a cross-section of $320 \text{ nm} \times 630 \text{ nm}$. The pump-to-signal detuning was less than 1 nm to ensure phase matching [7]. We measured the

dependence of conversion efficiency η on pump power (see Fig. S4(d)) and extracted the nonlinear coefficient γ by fitting the expression [8]

$$\eta = \frac{P_{\text{idler}}(L)}{P_{\text{signal}}(L)} = (\gamma P_{\text{pump}}(0) L_{\text{eff}})^2 \quad (\text{S1})$$

where $L_{\text{eff}} = (1 - e^{-\alpha L})/\alpha$ is the effective waveguide length and α the linear loss coefficient. The conversion efficiency was determined from the spectrum at the chip output. The pump power was estimated by subtracting the chip-to-fibre coupling loss from the fibre output power. We obtained an ultra-high nonlinear coefficient of $\sim 660 \text{ W}^{-1}\text{m}^{-1}$ and the derived material nonlinearity [9] of $\text{Al}_{0.17}\text{Ga}_{0.83}\text{As}$ is about $2.6 \times 10^{-17} \text{ m}^2\text{W}^{-1}$, which is similar to typical values previously reported [8,10,11].

SECTION III Frequency comb generation in AlGaAs-on-insulator microresonators

Fig. S5(a) shows the experimental setup for frequency comb generation in AlGaAsOI microresonators. A CW pump external cavity laser (ECL) (Ando AQ4321D), amplified by an erbium-doped fibre amplifier (EDFA, Amonics AEDFA), was passed through a tunable band-pass filter (3-dB bandwidth: 16 nm) to minimize amplified spontaneous emission noise. Two lensed fibres were used for fibre-to-chip coupling. The output spectrum was measured with an optical spectrum analyser (Yokogawa AQ6375). The temperature of the sample substrate was kept at 22.5°C to ensure stable coupling. The input and output power were always monitored by a power meter (Hewlett Packard 8153A, not shown) during the experiment.

Fig. S5. (a) Experimental setup for frequency comb generation in AlGaAsOI microresonators. (b) Evolution of Kerr frequency comb spectrum when the pump-resonance detuning is decreased. The pump power is kept the same (42 mW in this case) during the tuning.

Fig. S5(b) shows the evolution of frequency comb spectrum when the pump frequency is tuned into a resonance of the AlGaAsOI microresonator. Following primary sideband generation, sub comb lines were generated close to the primary sidebands and finally merged to form a frequency comb with the native line spacing (single-free spectral range), which shows a typical evolution as reported [12].

Fig. S6. Frequency comb generation performances of AlGaAsOI microresonators with different aluminium content. (a,b) Transmission spectra for resonances for the AlGaAsOI micro-resonator with 15% (a) and 17% (b) aluminium concentrations. (c,d) Measured output spectra when the pump is fine-tuned to a resonance for the micro-resonator with 15% (c) and 17% (d) aluminium concentration. The coupled pump powers are 30 mW and 7 mW in (c) and (d), respectively.

The bandgap of AlGaAs material can be engineered by changing the aluminium concentration during the epitaxial growth process. To investigate the impact of aluminium concentration on Kerr frequency comb generation, we fabricated two AlGaAsOI microresonators with similar Q but with different aluminium concentrations (15%, 17%). Fig. S6(a, b) show the transmission spectra for resonances around 1550 nm and Fig. S6(c, d) show the measured output spectra of these two devices when pump laser frequency is fine-tuned to the resonances of microresonators. As the bandgap [13] of $\text{Al}_{0.15}\text{Ga}_{0.85}\text{As}$ (1.61 eV) is not large enough to suppress two-photon absorption (TPA) efficiently [11], optical parametric oscillation (OPO) was not observed in the micro-resonator as shown in Fig. 6S(c) when a pump power of 30 mW is coupled into the device. However, the TPA-induced nonlinear loss can be reduced to a negligible level as the bandgap is increased with higher aluminium concentration [11]. Therefore, a slightly larger bandgap e.g. 1.64 eV ($\text{Al}_{0.17}\text{Ga}_{0.83}\text{As}$) results in a significant reduction of TPA and thus enables OPO in the micro-resonator as shown in Fig. S6(d) with a much lower pump power (7 mW).

The OPO threshold power is highly dependent on the quality factor and cavity length. Fig. S7(a) shows the estimated Q_L for microresonators with different cavity length based on [14]. For the current linear loss level (~ 1.4 dB/cm), the Q_L saturates at approximately 2×10^5 as the cavity length increases. In such case, the threshold power will increase linearly as the cavity length. Therefore, the threshold power will be approximately 25 mW if the FSR is reduced to 25 GHz. As shown in Fig. S7, improvement on linear loss will significantly boost the Q and thus dramatically decrease the OPO threshold power for resonators with small FSR

(<200 GHz). Once the linear loss is reduced to less than 1 dB/cm, the threshold power for a microresonator with 25-GHz FSR is reduced to the milliwatt level and if the loss is further reduced to less than 0.5 dB/cm, a sub-milliwatt threshold can be expected.

Fig. S7. Theoretical estimation of loaded Q (a) and OPO threshold power (b) for critical-coupled AlGaAsOI microresonators with different cavity lengths (different FSRs).

Reducing the waveguide sidewall-induced scattering is of critical importance for a low loss waveguide. Improvement of fabrication processes including high-resolution smooth patterning dry etching is highly desired to minimize the sidewall roughness [15]. In addition, reducing the field strength of the waveguide fundamental mode at the sidewall by using e.g. multi-mode waveguides is also efficient to lower the linear loss [16]. An over-cladding material with a higher index than SiO_2 (e.g. Al_2O_3 and TiO_2) can be applied to lower the index-contrast at the sidewall interface and thus the scattering [17-19]. Though it is very challenging to achieve low loss waveguide (<1 dB/cm) in high-index contrast platforms, ultra-low loss waveguides have been demonstrated in silicon-on-insulator (SOI) platform. For high- and weak-confinement SOI waveguides, the achieved lowest propagation losses are about ~ 0.7 dB/cm [20] and ~ 0.3 dB/cm [21,22], respectively. Therefore, we believe the similar performance can also be realized in the AlGaAsOI platform.

To further increase the Q for microresonators, a sophisticated bus-to-resonator coupler is required to reduce the coupler loss [23,24], bending losses might be reduced by design of adiabatic bends that curve smoothly instead of circular bends and a careful design for a junction between the bends and straight waveguides in race-track-shaped resonator is also favourable [14].

Fig. S8. Dispersion engineering of AlGaAsOI waveguide beyond $2 \mu\text{m}$.

Because of the wide transparency of the AlGaAs material, the frequency comb generation can also be realized in mid-infrared wavelengths as anomalous dispersion can be obtained in this wavelength range with larger dimension waveguides as shown in Fig. S8. This potentially allows for applications such as molecular spectroscopy [25].

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