Mid-Infrared Frequency Comb Generation Beyond 4 μm in Nanophotonic Lithium Niobate

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Abstract: We report frequency comb generation in the mid-IR beyond 4 μm in a dispersion-engineered periodically poled lithium niobate nano-waveguide through simultaneous second-harmonic generation, quadratic spectral broadening, and intra-pulse difference-frequency generation. © 2025 The Author(s)

In the past decade, various integrated photonic efforts towards mid-IR generation are driven by applications such as spectroscopy and biosensors, seeding development of different types of integrated sources ranging from broadband frequency combs to tunable sources [1, 3]. Recently, thin-film lithium niobate (TFLN) on sapphire became an emerging platform due to sapphire's wide transparency window, opening up opportunities for mid-IR wavelengths generation beyond 4 μm [4]. Spectral broadening mechanisms such as supercontinuum generation (SCG) has been studied, with designs focusing on efficient pump and second-harmonic (SH) phase matching [5]. Demonstrations of mid-IR wavelength generation via difference frequency generation (DFG) up to 4.2 μm has also been achieved in TFLN on sapphire substrates [2]. To exploit strong nonlinear interactions in confined TFLN waveguide modes over longer propagation lengths to efficiently generate mid-IR wavelengths, dispersion properties of targeted wavelengths must be put into considerations. In particular, poling periods for quasi-phase-matching and waveguide dispersions can be designed to simultaneously support both pump and SH signal phase-matching and mid-IR DFG, which can be dispersion engineered through targeting a flat group index over the bandwidth of interest. So far, the spectral coverage of such SCG processes in TFLN has been limited to below 4 μm. In this work, we theoretically show and experimentally demonstrate that while targeting low group velocity dispersion (GVD) and low group velocity mismatch (GVM) can lead to efficient SCG generation through saturated second-order processes [5], proper phase- and group velocity-matching of DFG processes offers a path to further extend the long-wavelength side of the generated spectrum towards 5 μm. We experimentally demonstrate such a properly dispersion- and phase-matching-engineered waveguide pumped by a femtosecond frequency comb source centered at around 2 μm and generating spectral content up to ~ 4.5 μm.

The concept of the spectral broadening mechanism is as follows. As shown in Fig.1(a-d), the input spectrum is first doubled through phase-matched second-harmonic generation (SHG, Fig.1(a)), which is followed by SCG through saturated $\chi^{(2)}$ processes (Fig.1(b)). Provided both phase- and group-velocity matching, the spectrally broadened pump and SH can lead to broadband DFG processes to further broaden the spectrum on the long side, as illustrated in Fig.1(c,d). The two scenarios for such DFG processes are: (i) the DFG between the spectral components of the input and the SH combs (DFG 1), and (ii) the DFG between the spectral components of the input comb (DFG 2), where the resulting mid-IR comb has zero f_{CEO}. To further illustrate effectiveness of simultaneous DFG phase-matching and group velocity engineering, we calculate and compare the group index of two different waveguide geometries: one with zero GVD and zero GVM between the pump and the SH in TFLN on silica for efficient saturated $\chi^{(2)}$ SCG [6] (Geometry 1), and another optimized for the proposed additional DFG processes in TFLN on sapphire (Geometry 2). While substrate material absorption and long-wavelength mode confinement significantly contribute to observation of mid-IR wavelengths in experimental results, here we simplify the theoretical analysis to be absorption-free in our simulations and focus on the proposed dispersion and group velocity engineering method. The results are shown in Fig.1(e), with Geometry 2 having a flatter n_g extending more into the longer wavelengths region. In Fig.2(f) and (g), we fixed the poling period to have perfect phase-matching between the pump and SH signal, then looked at potential DFG wavelengths between 3 μm and 5 μm and calculated the minimum GVM between all possible DFG phase-matched λ_1 and λ_2 pairs within the pump and signal bandwidth, slightly loosening DFG phase-matching condition to less than 0.5 rad/mm phase-mismatch. Evidently, Geometry 2 supports a much wider DFG phase-matching bandwidth with near-zero GVM values up to 4.5 μm, suggesting longer interaction lengths for DFG. Fig.1(h) and Fig.1(i) then show the DFG phase-matching curves plotted against λ_2 for Geometry 1 and Geometry 2, respectively, where zero crossings represent perfect phase-matching. While both geometries support perfect phase-matching ($\Delta k = 0$), Geometry 2 simultaneously has a wider DFG phase-matching bandwidth as well as near-zero GVM values up to 4.5 μm (Fig.1(g)). Finally, we proceed to nonlinear propagation simulations based on the single envelope equation, neglecting both material absorption and third-order (Kerr) nonlinear effects. As shown in Fig.1(j), for Geometry 1, spectral broadening is observed but with negligible spectral power above 3.2 μm even when the pump power is further increased, in agreement with our GVM analysis. In contrast, in Geometry 2, the phase-matched and group-velocity-matched DFG processes quickly extend the spectrum towards 5 μm, requiring much less pump power than Geometry 1.

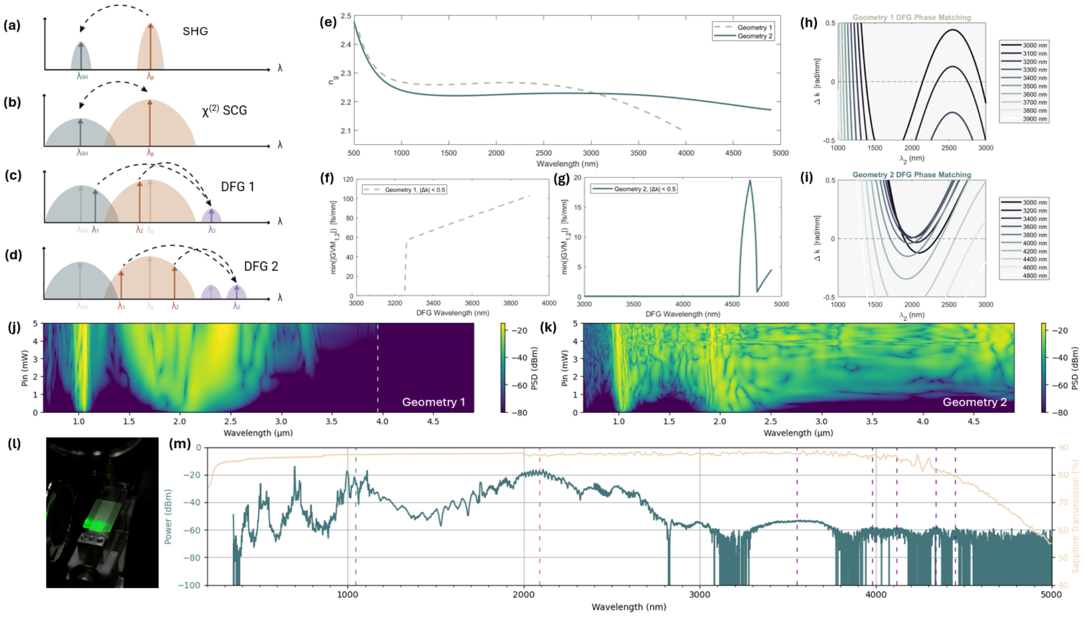

Fig. 1: **(a-d)** Schematic of the spectral broadening mechanism. **(e)** Group index comparison bewteen Geometry 1 and Geometry 2. **(f,g)** Minimum GVM between all possible DFG phase-matched λ_1 and λ_2 pairs. **(h,i)** DFG phase matching curves for Geometry 1 and Geometry 2, respectively. **(j,k)** Simulations of SCG at increasing on-chip pump powers for Geometry 1 (dashed line represents highest wavelength of supported mode) and Geometry 2, respectively. **(l)** Sample image. **(m)** Output spectra of DFG-SCG device with $\sim$ 539 mW input pump power off-chip.

We fabricated the device on a 975 nm TFLN on sapphire platform (NanoLN) via electron beam lithography and argon etching, with a top width of 3706 nm, etch depth of 516 nm, and poling period of 8.46 μm over 7 mm. This geometry supports fundamental TE mode up to $\sim$5 μm, a pump group velocity dispersion around -5 fs^2/mm, and a low but non-zero group velocity mismatch around 32 fs/mm between the pump and SH. The device is pumped by a femtosecond ($\sim$ 45 fs transform limited pulse width) optical parametric oscillator output centered at 2090 nm at 250 MHz repetition rate, with estimated total coupling loss of $\sim$ 23.5 dB. An image of the chip in the setup is shown in Fig.1(l). Experimental data with 2 μm pumping confirms DFG wavelengths in 3-4.5 μm range starting to appear above $\sim$250 mW of off-chip pump power, and the highest pump power ($\sim$539 mW) output spectra is shown in Fig.1(m), with DFG wavelengths centered around 3550 nm, 3980 nm, 4120 nm, 4340 nm, and 4450 nm, plotted along with the transmission spectrum of sapphire. We observe that this range of DFG spectral content is consistent with the near-zero GVM and DFG phase-matching curves presented in Fig.1(g) and Fig.1(i), suggesting good agreement between the dispersion engineering and experimental results.

In conclusion, we have demonstrated a new scheme for frequency comb generation in the mid-IR through simultanous SHG, SCG, and DFG process in a dispersion-engineered and quasi-phase matched waveguide in TFLN on sapphire. Purging the experimental setup can potentially enhance the mid-IR signal in regions of CO_2 absorptions, and beatnote measurements at the DFG wavelengths can provide more insight for finding which spectral contents from signal and/or pump combs are generating particular wavelengths. Further optimization of the design is possible through adjusting the quasi-phase matching as a function of length for more efficient DFG and higher powers on the long-side of the spectrum. Our results highlight a path for engineering the spectral coverage of on-chip broadband frequency combs in the mid-IR.

References

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