Broadband UV-Vis Frequency Combs from High-harmonic Generation in Quasi-phase-matched Waveguides

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Abstract:
We report efficient, phase-coherent high-harmonic generation in chirped periodically poled lithium niobate waveguides pumped with a watt-scale 3 μm frequency comb. Simulations support a mechanism of cascaded quadratic nonlinearity and provide insight into spectral optimization.

Ultraviolet-visible (UV-Vis) optical frequency combs are central to various metrology, spectroscopy, and sensing technologies [1], and ultra-broadband combs across the UV-Vis enable access to attosecond timescales at high-repetition rate [2]. High-order harmonic generation (HHG) is a leading method for coherently generating light of both short wavelength and short pulse duration, reaching the deep ultraviolet to as far as soft x-ray spectral regions and capable of sub-femtosecond brevity. Typically HHG is performed in atomic gas jets, but more recently HHG from solids has been demonstrated [3]. While most solid-state HHG work has been done with ~mJ pulses and thin crystals, optical waveguides present several opportunities for enhanced conversion efficiency in nJ-driven HHG [4] by providing wavelength-scale confinement in strongly nonlinear materials that may be microstructured to quasi-phase-match harmonic generation over a broad bandwidth. Gaining insight into the generation mechanisms in such structures is a key step toward tailoring harmonic emissions to suit applications.

Here, we present harmonic generation up to 9th order (333 nm) from chirped periodically poled lithium niobate waveguides pumped with phase-stabilized 3 μm, 100 fs pulses at 100 MHz repetition rate. We observe a mid-IR to UV-Vis conversion efficiency of about 10% amongst an overall 23% conversion of the fundamental to all harmonics. Numerical simulations based on the nonlinear analytic envelope equation [5, 6] are performed to account for the observed results in terms of cascaded-$\chi^{(2)}$ mechanisms. Despite the highly nonlinear HHG process, we also verify that the high harmonics inherit the coherence of the driving mid-IR comb.

In our experiments, mid-IR pulses are first derived from the 3 μm idler of the high-power optical parametric amplifier (OPA) described in [7], then coupled with 60% efficiency into five different zinc-oxide-doped lithium niobate waveguides [4], each with the same cross-section (15 × 16 μm) and length (25 mm) but different chirped

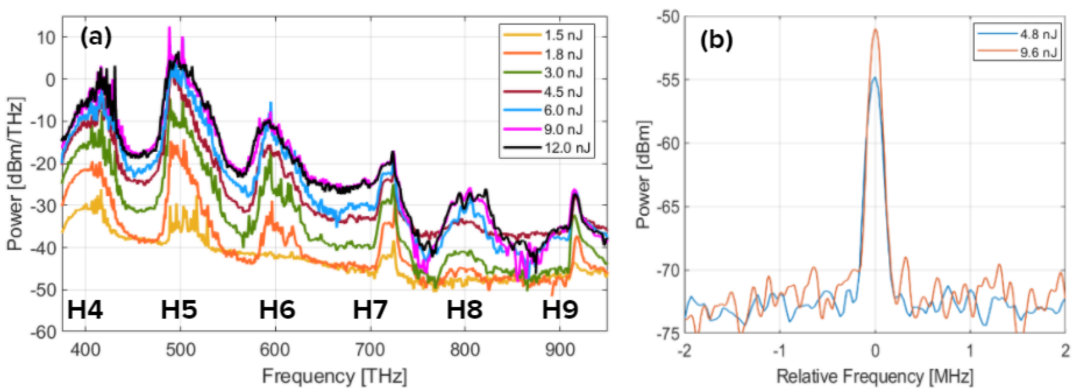

Fig. 1. **(a)** Measured spectra at various pump pulse energies in the waveguide. **(b)** Heterodyne beat notes (100 kHz resolution bandwidth) between the 6th-harmonic from the waveguide and second harmonic of the Yb:fiber pump comb used in the OPA, verifying the harmonic coherence and its preservation as power is scaled.

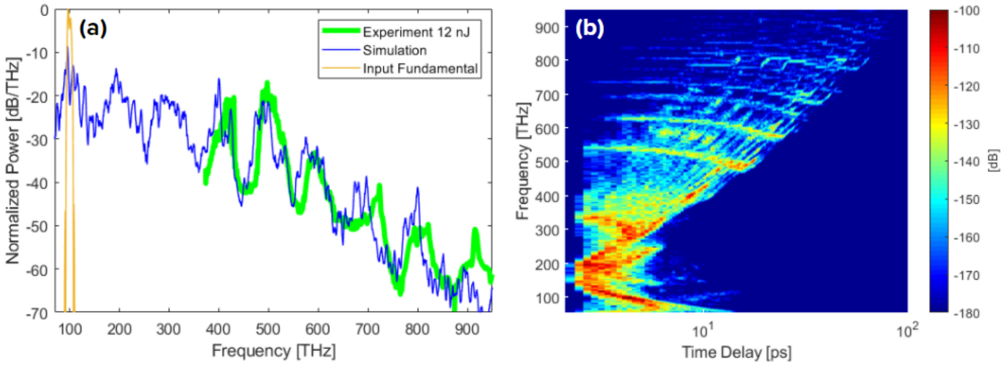

Fig. 2. **(a)** Overlay of simulated input pump (yellow), output (blue) and experiment (green). **(b)** Simulated spectrogram corresponding to the output spectrum in (a) (positive times lag the pump).

poling profile spanning periods from 20 to 30 μm. The mid-IR driver, with photon energy well below the band gap, enables high peak intensities in the waveguide $> 10^{11}$ W/cm^2 at 100 MHz repetition rate without damage.

Fig. 1(a) shows the measured absolute power spectral density for harmonics four through nine (H4-H9) at various pumping powers through a waveguide in the decreasing poling period direction. No harmonics are detected at 1.2 nJ pumping while all but H6 and H8 appear at 1.5 nJ, above which all harmonics scale with pumping power until they saturate around 9 nJ, indicating that increased pumping does not increase harmonic yield. The absence of 10th- and higher harmonics is attributed to above-bandgap (310 nm) absorption [8]. Testing other waveguides and directions produces all similar spectra except in some cases a suppressed 6th-harmonic. This suggests that while quasi-phase-matching is indeed important [4], the HHG efficiency appears not critically dependent on the shape of the poling profile but rather on its span. Fig. 1(b) shows, at two pumping powers, a free-running heterodyne beat note between the 6th-harmonic of the waveguide output and the second harmonic of the OPA 1 μm pump. The beat note is resolution-bandwidth limited at 100 kHz in both cases, implying excellent phase-coherence of the combs via HHG. In addition, we measured an integrated root-mean-square intensity noise of < 0.1 in a 1 nm bandwidth at the 7th-harmonic (428 nm) over a duration of 30 minutes, highlighting the stability of our experimental setup.

To better understand the mechanisms responsible for HHG, we model the process via 1D simulations accounting for second-order nonlinearity, dispersion, and self-steepening [5, 6]. Although a more sophisticated model could accommodate the dispersion of the nonlinear susceptibility and the multi-mode nature of the waveguide [9], this simpler model reproduces the relatively smooth measured spectrum and achieves reasonable agreement with the experiments when run with reduced pumping power. Fig. 2(a) show a representative example comparing simulated spectra with experimental data. Further, the simulation allows for detailed analysis of the HHG process, for example as shown in the spectrogram of Fig. 2(b). At the waveguide output, the harmonics lag the fundamental due to the material dispersion. Analysis of spectrograms at other points in the waveguides shows how the sum frequency generation of higher harmonics depends on the build-up of lower harmonics in a cascaded process, yet also that difference frequency generation among them is significant. We find that most harmonic generation occurs in regions of steep variation of the poling period profile, where successive phase-matching conditions can be rapidly sampled prior to temporal walk-off of participating fields. This detailed understanding can aid in optimization of the cascaded HHG process. For example, via optimizing the input pulse and poling period, custom tailoring of the spectrum to specific harmonics, or broadband quasi-continuum generation can be achieved.

References

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