Low-Harmonic Generation in Cascaded Thin-Film Lithium Niobate Waveguides

Tracy Sjaardema, Marcin Malinowski, Ashutosh Rao, and Sasan Fathpour

Herein, frequency conversion to the third- and fourth-order harmonics are demonstrated in ultracompact cascaded periodically poled thin-film lithium niobate (LN) waveguides on silicon substrates. Pumped at 1550 nm wavelength, the tightly confined waveguides produce strong third- (523 nm) and fourth-harmonic (386 nm) signals. Actively monitoring the periodic poling process on the LN devices renders these efficient frequency conversions. Measurements with a femtosecond pulsed laser source result in comparable power levels across the generated harmonics. A model that simulates harmonic generation from the pulsed source is presented and discussed. The model’s results are in general agreement with the experimentally observed processes.

1. Introduction

Nonlinear frequency conversion generates wavelengths of light that may not be conveniently available from conventional laser sources.[1] Integrating these sources can be particularly useful toward on-chip circuits and systems, as well as more efficient and smaller-footprint devices that operate at lower optical pump powers than bulk-crystal approaches. The small modal effective area of the integrated waveguides provides a high nonlinear overlap between the interacting modes. Thus, much attention has been given to integrated frequency conversion approaches, including harmonic generation,[2–11] frequency combs,[12–14] and even quantum-frequency conversion.[15,16]

With its high second-order nonlinearity ($\chi^{(2)}$) and broad transmission window, lithium niobate (LiNbO$_3$, LN) is a well-established material for frequency conversion.[17–19] Since the nonlinear conversion efficiency is inversely proportional to the nonlinear interaction area, an ultracompact (submicron-scale) waveguide that tightly confines the mode in the nonlinear material can offer high efficiencies.[3,20–22]

Conventional LN waveguides are formed through diffusion and proton-exchange techniques.[23,24] These low-index contrast waveguides suffer from large, weakly confined modes and thus lower conversion efficiencies. Optical waveguides, based on machined ridges in moderately thick LN films bonded to a carrier substrate, have been pursued in the past.[25,26] Thin-film LN has proven to be a more promising platform to address this issue for many nonlinear integrated devices.[4,17,27] Thin-film LN on silicon substrates is an especially promising platform as it offers waveguides with tightly confined modes and potentially compatible with the well-established field of silicon photonics.[21,28] Thin-film LN wafers are typically formed through ion implantation, bonding, and thermal slicing processes, where the device layer is less than one micron thickness, and is isolated from the substrate by a silicon dioxide bottom cladding layer.[29] LN on sapphire substrates is also another candidate for realizing tightly confined waveguides.[30] Both rib-loading and direct-etching approaches have been employed to form optical waveguides on the thin-film LN wafers.[2,21,28,31,32]

Sum-frequency generation (SFG) and second-harmonic generation (SHG) are two major examples of second order, $\chi^{(2)}$, nonlinear processes implemented through three-wave mixing.[1] Different methods are used for quasi-phase-matching (QPM) to overcome the momentum mismatch of the photons in these processes, periodically poled lithium niobate (PPLN) is the most common method.[33–35] PPLN has been used in bulk[36,38,39] and thin-film[3,20–22] platforms in the past.

Most recently, SHG with record-high efficiency of 4,600% W$^{-1}$ cm$^{-2}$ has been demonstrated in thin-film PPLN on Si, with a device length of 0.6 mm.[38] The devices as mentioned by Rao et al.[28] were so efficient that third-harmonic generation (THG) and fourth-harmonic generation (FHG) were observed in them. However, due to the high dispersion of thin-film LN waveguides, the periodicity of the grating in a single QPM segment (designed for SHG at 770 nm) was not optimized for THG and FHG. To optimize the higher-harmonic conversion processes, a second QPM segment designed to optimize the THG/FHG processes can be inserted and integrated with the first PPLN device. Such cascaded thin-film LN waveguides for low-harmonic generation is the topic of the present work.

More interestingly, these integrated PPLN segments for $\chi^{(2)}$ frequency conversion processes can be cascaded for generating the third and fourth harmonics. Conventional PPLN waveguides have been used to demonstrate cascaded third-order[40] and
higher-order\textsuperscript{[5]} harmonic generation. Additionally, THG is demonstrated through a cascaded process on LN-on-insulator (LNOI) microdisks\textsuperscript{[35]} and cascaded modal phase-matching.\textsuperscript{[38]} In this work, strong THG and FHG are demonstrated in dual-pitch QPM segments in ultracompact waveguides on thin-film PPLN-on-Si\textsuperscript{[39]}

The third harmonic (TH) can be generated by using SHG and SFG waveguide segments. The SHG segment generates the second harmonic (SH) of the input pump wavelength, and the SFG generates the TH from the sum of the pump and the SH. The fourth harmonic can be similarly generated by SHG segments, where the first segment doubles the pump frequency to generate the SH, and the second segment doubles the SH frequency to generate the fourth harmonic. As far as the transparency range of LN (0.35–5.2 μm) is not violated, other combinations of $\chi^{(2)}$ processes can also be added to reach even higher harmonics, for example, adding a third segment, quasi-phase-matched for SFG, to the circuits described earlier could attain fifth and sixth harmonics, respectively. These cascaded structures can also be tuned to reach a broad span of wavelengths.

Efficient THG and FHG can have potential applications in a host of integrated photonic applications. For example, the generated signals can be used as on-chip light sources at visible and at ultraviolet wavelength ranges. This can be more conveniently achieved by using available integrated InP-based semiconductor lasers, as opposed to the challenges of integrating III-nitride lasers on the employed platform, for example, silicon photonics. Another example application is novel integrated optical isolators based on efficient nonlinear processes such as difference-frequency generation (DFG)\textsuperscript{[40]} as well as SHG.\textsuperscript{[41]} In such isolators, it may be desirable that the generated idler is converted back to the original signal wavelength, hence efficient cascaded parametric processes are required.

### 2. Method

COMSOL simulations are run to find the optimal device dimensions, to ensure high mode overlap and tight confinement for the transverse-electric (TE) fundamental modes of the pump, SH, TH, and fourth harmonic. The effective index of each of these TE modes allows for calculating the poling period of each of the PPLN segments for optimal phase-matching. The spacing between the poling electrodes (8 μm) is chosen such that the spacing is wide enough to result in minimal interference of the electrodes with the modes, but narrow enough for the available applied electric field (limited by a 400 V power supply) to exceed the 21 kV mm\textsuperscript{−1}\textsuperscript{[42]} coercive field strength of LN necessary for poling the material. The final device design is shown in Figure 1.

Table 1 summarizes the dimensions of the parameters for each of the devices, designed for 1550 nm input light. A small gap (10 μm) is placed between the two PPLN segments to separate the segments enough to allow each segment to be poled separately. The duty cycle chosen for all of the metal electrode periods is 35%, which has been shown to give a 50% duty cycle for the inverted domains.\textsuperscript{[2]}

The devices are fabricated on 300 nm thin-film X-cut LN on silicon.\textsuperscript{[29]} The first processing step is formation of electrodes for poling. Due to the precision required in fabricating the small dimensions of these electrodes, patterning is performed using electron-beam lithography. After the lithography, the electrodes are formed by depositing thin layers of chrome and gold (10 and 100 nm thick, respectively) via electron-beam evaporation. A lift-off procedure is then performed in N-methyl-2-pyrrolidone heated to 120 °C to form the poling electrodes.

A second electron-beam lithography process is next used to pattern the waveguides. ZEP 520 A resist is used in both lithography steps, and the patterns are developed in ortho-xylene developer. The waveguides are formed through dry etching in

| Table 1. Device dimensions. | THG device | FHG device |
|-----------------------------|------------|------------|
| Input wavelength            | 1550 nm    | 1550 nm    |
| First-segment process       | SHG        | SHG        |
| First-segment length        | 800 μm     | 800 μm     |
| First-segment poling period | 2.83 μm    | 2.83 μm    |
| Wavelength after first segment | 775 nm  | 775 nm     |
| Second-segment process      | SFG        | SHG        |
| Second-segment length       | 1800 μm    | 1800 μm    |
| Second-segment poling period | 2.20 μm  | 1.33 μm    |
| Wavelength after second segment | 516.7 nm | 387.5 nm  |

\textsuperscript{[a]}FHG: first-harmonic generation; FSH: fourth-harmonic generation; SFG: Sum-frequency generation; SHG: second-harmonic generation; THG: third-harmonic generation.

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**Figure 1.** a) Waveguide cross section with labeled dimensions. b) Device schematic with labeled dimensions.
an argon plasma, with subsequent cleaning steps in an oxygen plasma and a dilute hydrofluoric acid (HF) solution (1:100 49% HF:H₂O). The waveguide facets are formed by dicing and polishing.

The devices are poled using the poling setup shown in Figure 2, consisting of a function generator (FG) to generate the pulses, an oscilloscope (OSC) to monitor the pulses, a high-voltage amplifier (HVA) to amplify the voltage pulses by a factor of 100, and two tungsten contact probes to connect to the electrodes on either side of the waveguide. The pulses generated by the FG are 15 ms square pulses with a 3.7 V amplitude and a repetition period of 5 s.

The devices are poled by employing an optically monitored iterative poling process, where the output of the device is monitored on an optical spectrum analyzer (OSA) before and during poling.[28] Before the devices are poled, a pulsed laser (with 70 fs pulse width at 80 MHz repetition rate, pulse energies up to 115 pJ, and a wavelength centered near 1550 nm) is lens-coupled into the device and out-coupled via a lensed fiber that leads to the OSA. At this stage, the OSA shows only the input pulse spectrum, confirming no harmonic generation is produced through unintentional phase-matching in the waveguide. Then, with the OSA continuously scanning, the first segment is poled. The output is monitored and as poling continues, the power of the SH increases. To avoid the possibility of overpoling the device and causing a decrease in conversion efficiency, the poling is stopped after the SH power begins to saturate. The same actively monitored process is repeated for all remaining PPLN segments. Figure 2 depicts this poling process, showing no harmonic generation before poling, and showing examples of the harmonics generated after poling each segment of a THG device.

3. Results and Discussion

After poling, the waveguide is first characterized with a tunable, continuous-wave (CW) source to find the conversion efficiencies of the SHG segments, using the setup shown in Figure 3. A tunable CW laser with a fiber-coupled output, centered at 1550 nm, is passed through a fiber-based polarization controller (PC) and coupled into the device via a lensed fiber. Light is coupled out of the device through another lensed fiber and impinged upon a silicon photodetector. While measuring the amount of light generated at the SH, the input laser wavelength is swept 100 nm to find the pump wavelength at which the generated SH is highest. This gives the pump wavelength that best satisfies the

![Figure 2](https://www.advancedsciencenews.com/)

*Figure 2. Poling setup and optical spectrum analyzer (OSA) results after poling each segment.*
phase-matching condition. This phase-matched wavelength is measured to be 1585.6 nm for the THG device, as shown in Figure 4, and 1553.2 nm for the FHG device.

The equation for calculating the conversion efficiency in %/W is given by $\eta = \frac{P_{2\omega}}{P_\omega}$, where $P_\omega$ is the power of the pump in the waveguide and $P_{2\omega}$ is the power of the SH in the waveguide. The accordingly measured SHG CW conversion efficiencies for the first segment of the THG and FHG devices are 3.84 and 2.56% W$^{-1}$, respectively, with segment lengths of 800 μm. It is noted that normalized efficiencies in short devices need careful deliberation of the propagation loss, as the quadratic dependence of output power versus length is strictly applicable to the low-loss regime. In addition, the e-beam stitching error in fabrication of the grating of longer PPLN devices can contribute to higher loss. Measurements on other devices fabricated in the same way as those shown here have shown insertion losses of 6 and 7 dB per facet, and propagation losses of 3 and 7 dB cm$^{-1}$, for pump and SHG wavelengths, respectively. These loss values are assumed to be the same for these devices.

CW measurements, even those at high powers through use of an erbium-doped fiber amplifier (EDFA), do not show higher-order harmonic generation. This is likely due to difficulty in achieving the precise phase-matching conditions. Therefore, a pulsed source is used to measure the higher-order harmonics. A mode-locked femtosecond pulsed laser source is used to measure the harmonic generation of the devices at multiple input powers. The laser output pulse specifications are 70 fs pulse width at 80 MHz repetition rate, pulse energies up to 115 pJ, and a wavelength centered near 1550 nm. The spectrum of the input pulse is shown in the inset of Figure 6. The setup for the pulsed-source measurement, shown in Figure 5, consists of the pulsed laser detailed earlier, a variable attenuator (ATT), a half-wave plate (HWP), and a polarizer (POL). The input light is coupled into the device with a 100× objective lens, and is out-coupled with a lensed fiber. This fiber is connected to a long-range OSA that detects wavelengths between 350 and 1700 nm. The variable ATT is used to adjust the energy of the input pulses, and the output sweep from the OSA is saved for each input pulse energy.

Figure 6 displays the OSA data for each input pulse energy, showing how the power in each harmonic varies with input pulse energy for the THG device. Figure 6 displays the cross sections for three of these input pulse energies. From the spectra for the maximum input pulse, it can be seen that the power level of the generated TH is comparable to that of the SH, indicating an efficient conversion process compared to single-grating PPLN devices (see Figure 4c in ref. [28]). However, the conversion efficiency of the data from a pulsed source cannot be extracted in the same straightforward way as for a CW input, due to complexities arising from the difference between the peak and average powers, the broad and spectrally rich bandwidth, and the phase components of the pulses.

Therefore, a simulation model is developed to analyze the nonlinear processes, and the conversion efficiency is extracted from the best fit to the experimentally measured data. The mathematical framework of the model is presented in the Supporting Information. The model solves the nonlinear Schrödinger equation for coupled modes with second-order nonlinearity for SHG and SFG. The model requires several inputs, including the effective indices, group velocity, group velocity dispersion, and overlap areas, that are determined through COMSOL mode simulations, and system losses that are measured experimentally. The input pulse used in the model is formed by taking the pulse spectrum in frequency domain from an OSA measurement of the pulse used in the experimental measurement, and converting the data into time domain. This ensures the pulse used for the simulation contains the same spectral components as the pulse used in the experimental measurements. Chirp is added to the pulse to produce a quadratic phase, and the chirp parameters are tuned such that the simulation output for a test case matches known results. In future works, frequency-resolved optical gating (FROG) measurements could be performed to more precisely obtain the spectral phase of the pulse.

The CW conversion efficiencies measured experimentally for the first segments of both devices are set as constants in the model. The simulated SHG spectra of the first segment for both the THG and FHG devices are comparable to the experimental results. The conversion efficiencies for the second segments are

Figure 3. Setup for measuring the conversion efficiency of the first (second-harmonic generation [SHG]) segment of the devices with a continuous-wave (CW) source.

Figure 4. CW phase-matching plot for SHG in the first segment (800 μm) of the third-harmonic generation (THG) device.

Figure 5. Setup for measuring the devices with a pulsed source.
returned by the model by fine-tuning simulation parameters, including group velocity, group velocity dispersion, overlap areas, and wavevector mismatch to achieve the best fit between the simulated and measured results.

**Figure 7** compares the experimentally measured spectra to the results returned by the simulation for the THG device, demonstrating agreement between them. Extracting the conversion efficiency from the model gives $28.8 \pm 8.5\%$ W$^{-1}$ for the SFG conversion process for generating the TH. The uncertainty in the conversion efficiency comes from adding a $\pm 5\%$ change to simulation parameters and extracting the resulting range of conversion efficiencies. This is due to uncertainty in the precise phase-matching conditions in the sample, limitations of the simulation process, and subjectivity in determining the coupling coefficient that best fits the data. The output is evidently very sensitive to small changes in many of the input parameters.

The linewidth of the measured TH and fourth harmonic is broader than the initial model predicted (not shown here), thus measures were taken to improve the model. Particularly, possible nonuniformity in film thickness along the waveguide would gradually change the phase-matching condition, causing slight shifts in the generated wavelength. This phenomenon would broaden the TH and fourth harmonic peaks. This fabrication nonuniformity is implemented in the model by adding a linear
ramp to the phase-matching condition along the length of the device, around the center wavelength. This effectively broadens the TH and fourth harmonic peaks, and, as can be seen in Figure 7, achieves a good fit to the experimental measurements.

The FHG device is characterized similarly. Figure 8 images the OSA traces for each of the input pulse energies, showing how the power in each harmonic grows with increasing input pulse energy for the FHG device. Figure 8 displays the OSA traces for several of these input pulse energies.

From the spectra for the maximum input pulse in Figure 7, it can be seen that the power level of the generated fourth harmonic is comparable to that of the SH, again indicating efficient conversion compared with the single-PPLN devices as mentioned by Rao et al. The model is rerun for the FHG data in a similar manner to that for the THG process.

Extracting the conversion efficiency from the simulation gives 9.91 ± 1.2% W⁻¹ for the SHG-based fourth harmonic conversion process, which is about a factor of three smaller than the SFG process in the THG device. As with the THG case, the uncertainty in the conversion efficiency comes from adding a ±5% change to simulation parameters and extracting the resulting range of conversion efficiencies.

Figure 9 compares the experimentally measured spectra to the simulation results. The fourth harmonic peak from the model is narrower than the measured peak, even after adding the aforementioned nonuniformity to the model. This discrepancy

Figure 8. a) Harmonic generation (optimized for fourth-harmonic generation [FHG]) as a function of input pulse energy. b) Harmonic generation measured in decibel-milliwatts (dBm) for multiple input pulse energies.

Figure 9. Comparison of the measured and simulated spectra for (from left to right) the pump, SH, and fourth harmonic.
is larger than that for the THG, and is not completely understood. Mode simulations show a drastic difference between the values of the overlap integrals of the fundamental mode of the SH with the fundamental mode of the fourth harmonic and the third-order mode of the fourth harmonic. Thus, the discrepancy is not from the occlusion of higher-order modes from the model.

As is evident in Figure 5 and 7, a small amount of fourth harmonic is generated by the THG device, and a small amount of TH is generated by the FHG device. Experimentally, this is likely due to cascaded, non-optimized SHG and SFG phase-matching, respectively, as was observed in previous, similar single-segment PPLN.[28] The amplitudes of these generated harmonics are, however, very small relative to the other harmonics, and cannot be the source of the discrepancy for FHG in Figure 9.

4. Conclusion

In conclusion, this work shows that by cascading two $\chi^{(2)}$ processes, third- and fourth-order harmonics can be generated, providing a link between telecommunication wavelengths to visible and ultraviolet wavelengths. These harmonics are generated by pumping ultracompact, thin-film PPLN-on-Si devices with 1550 nm CW and pulsed laser sources. Pulsed input measurements demonstrate harmonic generation from these cascaded PPLN devices to the fourth-order harmonic. Modeling the pulsed-input harmonic generation processes allows for the extraction of the conversion efficiencies for the higher harmonics, which are $28.8 \pm 8.5\%$ and $9.91 \pm 1.2\%$ W$^{-1}$ for the SFG and SHG processes that produce the third- and fourth harmonics, respectively. Cascading more segments could enable the generation of higher-order harmonics, and the range could be further extended by designing the waveguides to support inputs of longer wavelengths.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

harmonic generation, integrated optics & photonics, nonlinear optics, silicon photonics, thin-film lithium niobate

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