# Lithium Niobate Nonlinear Nanophotonics

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Lithium Niobate Nonlinear Nanophotonics

A DISSERTATION PRESENTED
BY
CHENG WANG
TO
THE SCHOOL OF ENGINEERING AND APPLIED SCIENCES
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY
IN THE SUBJECT OF
ELECTRICAL ENGINEERING

HARVARD UNIVERSITY
CAMBRIDGE, MASSACHUSETTS
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ABSTRACT

Lithium niobate (LiNbO$_3$) is by far the most widely used second order nonlinear optical material due to its high $\chi^{(2)}$ coefficient, wide transparency window (400 nm – 5 µm) and relatively high refractive index (~ 2.2). In nonlinear wavelength conversion, periodically poled lithium niobate is a standard platform for generating classical and quantum light sources, as well as frequency combs. In optical communications, lithium niobate is the material of choice for electro-optic modulators that require large data bandwidth, high signal integrity and low insertion loss. Conventional lithium niobate devices, however, achieve waveguiding by local perturbation of the crystal (e.g. ion diffusion) with low refractive index contrast, large mode size, and reduced nonlinear interaction strength. As a result, these devices are large, expensive, and require lots of power to operate, posing increasing challenges to modern optics applications that demand scalable solutions with low cost and low power consumption.

This thesis describes the realization of integrated lithium niobate photonics by combining the superior material properties of lithium niobate and top-down nanofabrication capability. By utilizing lithium niobate thin films (300 ~ 700 nm thick) bonded on top of silicon dioxide substrate, wavelength scale optical waveguides and cavities with excellent...
light confinement and strong nonlinear interactions can be realized. In Chapter 2, we first describe the optimization of nanofabrication approaches, in particular the dry etching process, that are capable of delivering on-chip photonic resonators with high quality factors (> 100,000).

Next, we show that the optimized thin-film lithium niobate platform can be used for versatile on-chip nonlinear optics applications. In Chapter 3, we propose and experimentally realize two distinct phase matching schemes for second harmonic generation in lithium niobate nanowaveguides, i.e. modal phase matching in uniform waveguides and quasi-phase matching in periodically grooved waveguides. In Chapter 4, we utilize a hybrid system consisting of amorphous silicon phase gradient metasurfaces and lithium niobate waveguides to control the light wave propagation properties inside the waveguides. Based on this platform, we show both efficient linear TE-TM mode conversion and phase-matching-free second harmonic generation. In Chapter 5, we show that by shrinking down the waveguide dimensions, lithium niobate electro-optic modulators can be brought into a new design paradigm which allows for much higher data bandwidth and lower driving voltage than their bulk counterparts. In Chapter 6, we present the design of photonic crystal nanogroove cavities with high quality factor and small modal volume that are promising for ultra-compact on-chip switches and nonlinear wavelength converters.
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Chapter 1

Introduction

In conventional (i.e. linear) optics, each optical element, including lenses, beam splitters and wave plates, can be treated as a linear system. This is because the underlining Maxwell’s Equations and corresponding constitutive relations are all linear functions. In particular, the light induced polarization ($P$) in a linear material is proportional to the applied electric field ($E$), i.e. $P = \varepsilon_0 \chi^{(1)} E$, where $\varepsilon_0$ is the permittivity in free space and $\chi^{(1)}$ is the linear susceptibility. As a result, the system output frequency (or color) follows whatever the input frequency is, without generating new frequencies. This can be unsatisfactory for certain applications that require the generation of new colors, or the manipulation of one photon with another photon.

Fortunately, the linear constitutive relations are only valid when the applied field strength is much smaller than the intrinsic atomic field strength. The nonlinear
polarization terms come into play in the cases of high optical power and/or strong field localization. The full field-induced polarization expression can be written as:

\[ P = \varepsilon_0 \left( \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots \right) = P^{(1)} + P^{(2)} + P^{(3)} + \ldots \]  

where \( \chi^{(1)} \) and \( \chi^{(2)} \) are the second- and third-order nonlinear susceptibilities, \( P^{(2)} \) and \( P^{(3)} \) are the second- and third-order nonlinear polarizations \( (1) \).

The leading term in Equation [1.1], or the \( \chi^{(2)} \) process, mixes the frequencies of two input signals \( (\omega_1, \omega_2) \) and creates a nonlinear polarization at a new frequency \( \omega_3 = \omega_1 \pm \omega_2 \). In the photon energy point of view, two photons \( (\omega_1, \omega_2) \) are annihilated in the process and a new photon \( (\omega_3) \) with is generated, where the total energy is conserved. Depending on the choice of \( \omega_1 \) and \( \omega_2 \), different applications can be realized using the \( \chi^{(2)} \) effect. For example, when the input frequencies are degenerate, frequency doubling of the input light can be achieved (second harmonic generation, SHG). The reverse process of SHG (spontaneous parametric down conversion, SPDC) is a major source for quantum entangled photon pairs.

Another example is electro-optic (EO, Pockels) effect, where \( \omega_2 \) is a static or low frequency \( (\omega_2 \ll \omega_1) \) electric signal, and the nonlinear process induces an additional polarization that oscillates at \( \omega_2 \), effectively modulating the refractive index. Typical \( \chi^{(2)} \) processes are listed in Table 1.1.
### Table 1.1 Second order nonlinear optical processes (1)

<table>
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<th>$\chi^{(2)}$</th>
<th>Process</th>
<th>Description</th>
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<tr>
<td>$\chi^{(2)}(2\omega_1; \omega_1, \omega_1)$</td>
<td>Second harmonic generation (SHG)</td>
<td>Input signal at $\omega_1$ generates frequency doubled output</td>
</tr>
<tr>
<td>$\chi^{(2)}(\omega_3; \omega_1, \pm\omega_2)$</td>
<td>Sum/difference frequency generation (SFG/DFG)</td>
<td>Input signals at $\omega_1$ and $\omega_2$ generate output at $\omega_1 \pm \omega_2$</td>
</tr>
<tr>
<td>$\chi^{(2)}(\omega_1; \omega_1, 0)$</td>
<td>Electro-optic (EO) effect</td>
<td>DC electric field modulates the refractive index at optical frequency $\omega_1$</td>
</tr>
<tr>
<td>$\chi^{(2)}(0; \omega_1, -\omega_1)$</td>
<td>Optical rectification</td>
<td>Optical signal generates DC electric field</td>
</tr>
</tbody>
</table>

1.1 Material Properties of Lithium Niobate

Due to the constraints by symmetry, the $\chi^{(2)}$ coefficients vanish in all centrosymmetric crystals or amorphous materials, including silicon (Si), silicon dioxide (SiO$_2$), silicon nitride (SiN), diamond (C) and most metals. Since the discovery of optical nonlinearity in the 60s, lithium niobate (LiNbO$_3$, or LN) has been the most widely used $\chi^{(2)}$ material, with applications ranging from nonlinear wavelength conversion for classical and quantum light source (1), optical modulators for data communications (2), as well as surface acoustic wave (SAW) based electronic components for mobile phone industry (3).
LN holds many favorable material properties in terms of both nonlinear and linear optics. The largest $\chi^{(2)}$ tensor component in LN is aligned diagonally ($\chi^{(2)_{zzz}}$), and is large for both optical wavelength conversion (known as $d_{33}$) and EO modulation ($r_{33}$). As a linear optical material, LN has a relatively large refractive index ($n_o = 2.21, n_e = 2.14$, at 1550 nm), and is highly transparent for wavelengths from 400 nm (blue) to 5 µm (mid-infrared), with an OH- absorption peak at 2.87 µm (4). In comparison, alternative $\chi^{(2)}$ materials exhibit either lower refractive indices (BBO, KTP, KDP), or nonlinear susceptibilities that are reduced (AlN, SiC, GaN) or non-diagonal (GaAs, GaP). A comparison of important nonlinear optical properties among representative $\chi^{(2)}$ materials is shown in Table 1.2.
Table 1.2 Optical properties of representative $\chi^{(2)}$ materials (~ 1500 nm wavelength)

<table>
<thead>
<tr>
<th>Material</th>
<th>Largest $d$ coefficient</th>
<th>Largest $r$ coefficient</th>
<th>Refractive index</th>
<th>Transparency window</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiNbO$_3$ (4)</td>
<td>$d_{33} = 27$ pm/V</td>
<td>$r_{33} = 27$ pm/V</td>
<td>~ 2.2</td>
<td>400 nm – 5 μm</td>
</tr>
<tr>
<td>BBO (4)</td>
<td>$d_{22} = 1.9$ pm/V</td>
<td>$r_{51} = 2.1$ pm/V</td>
<td>~ 1.6</td>
<td>200 nm – 2.6 μm</td>
</tr>
<tr>
<td>KTP (4)</td>
<td>$d_{33} = 14.6$ pm/V</td>
<td>$r_{33} = 35$ pm/V</td>
<td>~ 1.8</td>
<td>350 nm – 4.5 μm</td>
</tr>
<tr>
<td>GaAs (4)</td>
<td>$d_{36} = 119$ pm/V</td>
<td>$r_{41} = 1.53$ pm/V</td>
<td>~ 3.4</td>
<td>1.1 – 17 μm</td>
</tr>
<tr>
<td>AlN (5, 6)</td>
<td>$d_{33} = 4.7 \pm 3$ pm/V</td>
<td>$r_{33} \sim 1$ pm/V</td>
<td>~ 2.1</td>
<td>220 nm – 13.6 μm</td>
</tr>
<tr>
<td>GaN (7, 8)</td>
<td>$d_{33} \sim 20$ pm/V</td>
<td>$r_{33} = 1.91 \pm 0.35$</td>
<td>~ 2.3</td>
<td>365 nm – 13.6 μm</td>
</tr>
<tr>
<td>SiC (9-11)</td>
<td>$d_{33} \sim 10$ pm/V</td>
<td>$r_{33} = 2.7 \pm 0.5$</td>
<td>~ 2.5</td>
<td>&gt; 500 nm</td>
</tr>
</tbody>
</table>

1.2 Conventional Lithium Niobate Devices

Efficient guiding of light waves requires a refractive index contrast between the waveguide core and cladding materials. In conventional LN nonlinear optical devices and EO modulators, this is achieved by metal in-diffusion (e.g. Ti) or ion exchange (e.g. proton) in the desired waveguide regions, which locally perturbs the LN crystal and induces a minor refractive index change ($\Delta n \sim 0.02$) (12). The low index contrast results in weak light confinement, large mode areas (~ 10 μm × 10 μm) and bending radii (~ 10 cm) (Figure 1.1 left). Because the $\chi^{(2)}$ nonlinear
optical processes are intrinsically weak, and its relative strength is proportional to the optical intensity, conventional LN devices are relatively low efficient, cannot be densely integrated or mass produced.

On the other hand, recent years have seen tremendous progress in the field of nonlinear nanophotonics, realized in various material platforms including SiO\(_2\) (13, 14), Si (15-17), SiN (18, 19), GaN (20), GaP (21), AlN (22), GaAs (23), SiC (24) and diamond (25, 26). These nonlinear devices are usually fabricated from sub-micron thin films and possess high refractive index contrast (\(\Delta n > 0.5\)). The sub-wavelength light confinement results in strong photon-photon interactions and highly efficient nonlinear optical processes, including second harmonic generation (20-24), frequency comb generation (13-15, 18, 19, 25), photon pair generation (17), and Raman lasing (16, 26).

A fully integrated LN nonlinear optical platform, with high refractive index contrast and sub-wavelength light confinement (Figure 1.1 right), could overcome the limitations of conventional LN devices and enable efficient, low-cost and highly-integrated nonlinear optical systems.
1.3 Lithium Niobate on Insulator (LNOI)

In order to realize nanophotonic LN devices from a 3D bulk crystal, a two-step miniaturization process is needed (Figure 1.2). The first step is to fabricate LN thin films (3D \( \rightarrow \) 2D) with submicron thickness that are imbedded in a lower-index material (e.g. SiO\(_2\)). The second step further transforms the 2D slab into a 1D waveguide with sub-wavelength light confinement in both vertical and lateral dimensions.
Figure 1.2 Schematic of the two-step miniaturization process to realize nanophotonic LN devices from a 3D bulk crystal.

Lithium niobate on insulator (LNOI) has recently emerged as a promising solution for the first miniaturization step (27). Using a manufacture process similar to the preparation of silicon on insulator (SOI), single crystal lithium niobate thin films (< 1 µm thick) bonded on top of SiO2 are now commercially available (Figure 1.3). LNOI platform is the building block from which the various photonic devices described in the following chapters are fabricated.

Figure 1.3 Fabrication procedure of lithium niobate on insulator (LNOI) using a “smart cut” process (27).
1.4 Outline of the Thesis

The goal of this thesis is to describe an integrated LN nanophotonic platform that combines the unique nonlinear material properties of LN with the superior light confinement in wavelength scale optical waveguides and cavities.

**Chapter 2** describes the fabrication of high-quality nanophotonic devices in LN (step 2 in Figure 1.2). Using a top-down lithographical process, micro-disk resonators with optical quality factors > 100,000 are demonstrated. To test the material properties of the post-fabrication LN thin films, nonlinear wavelength conversion is shown and optical bistability is analyzed.

**Chapter 3** introduces two distinct phase matching techniques for SHG in sub-wavelength LN waveguides. Both modal phase matching in uniform waveguides and quasi-phase matching in periodically grooved waveguides are theoretically proposed and experimentally demonstrated, with normalized conversion efficiency as high as 41% W⁻¹cm².

**Chapter 4** describes the integration of LN waveguides and nanophotonic antenna arrays to manipulate the light wave evolution in waveguides. An efficient and low-loss linear optical mode converter (TE₁₁ → TM₁₁) is demonstrated. Based on
the same platform, a phase-matching-free SHG process is shown, with nonlinear generated signal monotonically accumulating over many coherent lengths.

**Chapter 5** describes a nanophotonic electro-optic platform that would allow for highly-efficient, ultra-compact and mass-producible modulators for future optical links. Both resonator-based and Mach-Zehnder interferometer (MZI) schemes are experimentally realized, with data operation rate as high as 40 Gbps.

**Chapter 6** describes the design of a photonic crystal nano-groove cavity in LN with high quality factor and small mode volume, promising for ultra-compact on-chip switches and efficient wavelength conversions.

**Chapter 7** summarizes the thesis and gives outlook towards a fully integrated LN photonic platform.
Chapter 2

High-Quality Nanophotonic Structures in Lithium Niobate

Owing to the recent development of LN thin film fabrication techniques via crystal ion slicing (27, 28), various micro- and nano-photonic elements have been realized in recent years, including microring resonators (29-31), microdisk resonators (32-34), photonic crystal cavities (35), and micro-waveguides (36-40). On-chip EO modulation (29-31, 34) and SHG (31, 35, 37-39) have been demonstrated in these devices. However, nanostructuring of LN, in particular its dry etching, is challenging. The fluorine (F) based plasma that is conventionally used to etch LN produces non-volatile etching product (LiF, melting temperature > 800 °C), which is redeposited on the etched surfaces and sidewalls, increasing surface roughness and reducing the etching rate (41). An unoptimized etching recipe usually results in rough sidewalls, large scattering loss and fairly low optical quality (Q) factors.
[4,000 in Ref. (29)]. In 2012, Wang et al. proposed that, by heating the microdisks close to its melting temperature after device fabrication, local melting on the disk edge could smoothen the surface through surface tension reshape, and the Q factors are subsequently increased to 26,000 (32). More recently, Q factors as high as 250,000 were obtained using femtosecond laser ablation followed by two-step focused ion beam (FIB) milling (33). While these results are promising, they utilize non-standard fabrication techniques that cannot be easily scaled up. Alternative hetero-structure approaches that circumvent the etching of LN have also been explored (30, 31, 40), but the device performance is compromised by the reduced overlap between optical fields and LN’s nonlinearity.

In this chapter, we demonstrate high-Q LN optical microdisk resonators fabricated by simple, robust and standard nanofabrication techniques, which require no post-fabrication processing and material treatment. These resonators operate at both visible and near infrared (NIR) wavelengths, featuring Q-factors as high as $1.02 \times 10^5$. A commercially available LN on insulator (LNOI) substrate, prepared via smart-cut techniques, was used to realize our resonators, which were subsequently fabricated by electron-beam lithography (EBL) and an optimized argon plasma etching process, which yield smooth sidewalls and result in high-
Qs. As a proof of concept, we demonstrate on-chip SHG in our devices, with an internal conversion efficiency of 10.9%W⁻¹.

2.1 Fabrication of LN Microdisk Resonators

The fabrication procedure used to realize our devices is summarized in Figure 2.1(a). LNOI substrates (by NANOLN) with a 400 nm thick LN device layer on top of 1 μm thick buried silicon dioxide layer, further supported on a single-crystal LN substrate, were used. Patterning of microdisk structures started with deposition of a 15 nm titanium layer on the LNOI substrate by electron beam evaporation. The titanium layer served two purposes: to promote resist adhesion and to provide conduction during subsequent EBL. Hydrogen silsesquioxane (HSQ) based negative-tone electron-beam resist (FOX®-16 by Dow Corning) was spin-coated on top of the titanium-coated LNOI substrate and patterned using EBL (Elionix ELS-F125). Resulting HSQ patterns were approximately 600 nm in height and used as an etch mask to define the LN microdisks. A second blanket electron beam exposure of the resist after development was used to increase the mask hardness, thus enhancing the etching selectivity. LN was then dry etched with argon plasma (Ar⁺) in an electron-cyclotron resonance (ECR) reactive ion etching
(RIE) system (NEXX Cirrus 150). A relatively high RF power (200 W) was used to enable DC bias of ~ 180 V that accelerates Ar+ ions towards the substrate, resulting in physical etching of the LN device layer. A typical etch rate is estimated to be ~ 30 nm/min. Approximately 300 nm of LN could be etched using this approach (leaving ~ 100 nm thick unetched LN layer behind), before significant mask erosion, which could result in pattern distortion and increased surface roughness. Following plasma etching, the remaining HSQ was removed by 2 min of wet etching in 5:1 buffered oxide etch (BOE). While the resulting ridge-like microdisks already supported optical modes, they were hard to be characterized using a fiber taper. To fabricate suspended LN microdisks on top of oxide pedestals, the remaining 100 nm thick LN layer was removed by an additional plasma etching step using the same ECR-RIE condition as before, but this time without any mask. Finally, the devices were undercut in 5:1 BOE (for 5 min) resulting in free-standing microdisks supported by a silica pillar.
Figure 2.1 Fabricated LN microdisk resonators. (a) Fabrication procedure. (b) Representative SEM image of a suspended 28 µm diameter LN microdisk supported by a silica pedestal on top of a LN substrate. A magnified view of the microdisk edge reveals smooth sidewalls. All SEM images are taken at a 45° angle.
Representative scanning electron microscope (SEM) images of a final LN optical resonator, with approximately 300 nm thickness, supported by 1 µm thick silica pedestal, are displayed in Figure 2.1(b). Since the argon plasma does not bombard the surface perpendicularly, sidewalls in our devices usually make an angle between 35° and 40° with respect to substrate normal direction. Nonetheless, the etched sidewalls are quite smooth with few noticeable rough sites under SEM inspection. Further characterization of other similarly etched samples using an atomic force microscope (AFM) shows the sidewall roughness to be less than 5 nm (RMS). Together with the fact that the top and bottom surfaces of our LN device layer have been polished to a surface roughness less than 1 nm (RMS), scattering loss is expected to be low, resulting in high optical Q-factors discussed next.

2.2 Microdisk Transmission Spectra in Telecom Wavelengths

LN microdisks were initially characterized in the telecom wavelength range, with a representative transmission spectrum collected from a 28 µm diameter disk shown in Figure 2.2(a). A silica fiber taper (42-44) was used as a local evanescent probe to couple light into and out of our microdisk resonators. Fiber tapers were produced by flame anneal and pulling from commercial single mode telecom
fibers (SMF-28, Dow Corning), resulting in a final diameter of ~ 1 μm. The pulled fiber taper was subsequently mounted in a U-shape and spliced into an optical set-up. The self-tension of the taper region makes it possible to be brought into close proximity to the desired device via motorized stages. The fiber taper is usually in physical contact with either top or side of the disk resonator, as is shown in the inset of Figure 2.2(a). Light from the tunable lasers (Santec TSL-510, tuning range from 1480 to 1680 nm) were sent into the tapered fiber after an inline fiber polarizer, and collected by a high gain InGaAs detector (EO Systems, IGA1.9-010-H). From the spectrum, three different sets of high Q-factor resonant modes are identified and displayed with different colors, each with similar free spectral range (FSR) near 13 nm. Numerical simulations from a finite element mode solver (COMSOL) indicate that these resonances correspond to transverse electric (TE)-like modes with the lowest three radial orders, all with negligible radiation loss. Each resonant dip is labeled as TE$_{mn}$, where m, n are the azimuthal and radial mode numbers respectively. Coupling strength to each mode can vary depending on the fiber position, resulting in different transmission dips for different modes. According to our modeling results in the telecom wavelength range, transverse magnetic (TM)-like modes are not supported in our devices since the LN device
layer is too thin. Figure 2.2(b)–(g) show high resolution transmission spectra corresponding to the three different radial mode orders, together with simulated cross-sectional mode profiles. Lorentzian fits to the resonances yield Q-factor estimates for the first, second and third order radial modes of $1.02 \times 10^5$, $8.8 \times 10^4$ and $4.4 \times 10^4$ respectively. The other distinguishable modes with lower Q-factors correspond to even higher radial orders. A large number of resonator modes from many devices were characterized, with different fiber-positions, and we find that nearly all devices support modes featuring Q-factors exceeding $5 \times 10^4$. 
Figure 2.2 Resonance spectra and simulation results in telecom wavelengths. (a) A representative transmission spectrum collected from a 28 µm diameter disk revealing several sets of resonances, indicated by color coding. Inset shows the optical micrograph of tapered fiber coupling on top of the microdisk resonator. (b-d) High-resolution views of representative resonance dips for each radial mode and their corresponding Lorentzian fits (black curves). (e-g) Finite element mode simulation of the three radial order modes shown in (b-d), indicating TE-like modes. Electric fields along radial direction (Er) are shown, labeled with mode numbers and effective indices. Dashed lines show the outlines of the device cross section.
2.3 Microdisk Transmission Spectra in Visible Wavelengths

LN microdisk resonator performance was further characterized in the visible and near-infrared (NIR) wavelength range ($\lambda < 1 \mu m$). To ensure relatively high modal overlap between the fiber and the microdisk modes for lower wavelengths, fiber tapers with a diameter of ~ 500 to 700 nm are needed. This time the fiber tapers were manufactured by a two-step hydrofluoric acid (HF) wet etching method (45), including ~ 30 minutes of concentrated HF etching, followed by ~ 30 to 45 minutes 5:1 BOE etching. A thin layer of o-xylene was covered on top of the HF solution in order to promote taper formation via the oil/water interface meniscus. Light from a super-continuum source (EXW-4, NKT Photonics) was delivered to the resonators using fiber taper coupling, and the transmission spectrum was collected with an optical spectrum analyzer (OSA, HP 70950B, minimum resolution bandwidth of 0.08 nm). Figure 2.3(a) displays the transmission spectrum of a 30 µm diameter microdisk resonator in the 770 to 780 nm wavelength range. At these wavelengths, the microdisk supports large numbers of TE and TM polarized optical modes, and within the FSR of the fundamental TE mode (~ 6 nm) as many as 16 different modes can be identified. The high density
of optical modes at these wavelengths is desirable for SHG experiments (discussed next). We note that the line widths of most of the resonances in Figure 2.3(a) are limited by the spectral resolution of our OSA, thus giving only a lower bound of their Q-factors. To obtain better estimates of the Q-factor values, we further characterized our device Q-factors at visible wavelengths by a tunable red laser (New Focus Velocity TLB 6304 laser, coarse tuning range of 634.8 to 638.9 nm and fine tuning range of 70 pm) and a visible band photodetector (New Focus 1801). Figure 2.3(b) shows the collected transmission spectra near 637 nm from the same microdisk resonator as in Figure 2.3(a). The fine tuning mode of the laser was used to accurately measure the Q-factors of cavity modes supported by the LN microdisk resonators within the ~ 4 nm coarse laser tuning range. High resolution spectra of two selected resonance dips with relatively high Q’s are shown in Figure 2.3(c-d). The spectra follow Fano-like shapes, likely resulting from interference between light coupling into the microdisk and light reflected from the contact region between the fiber taper and the microdisk (46), or from avoided crossing with other modes that are close in resonance wavelengths. The fine tuning resonance spectra were fitted into a Fano profile (47), with the relation:
\[
F(\lambda) = a_1 - a_2 \cdot \frac{\left(q + \frac{2(\lambda - \lambda_0)}{\gamma}\right)^2}{1 + \left(\frac{2(\lambda - \lambda_0)}{\gamma}\right)^2}
\]

[2.1]

where \(\gamma\) is the linewidth of the resonance, \(\lambda_0\) is the resonance wavelength, \(q\) is the Fano parameter related to the asymmetric line shape, \(a_1\) and \(a_2\) show the baseline and dip depth respectively. Q-factor can be calculated as \(\lambda_0/\gamma\).

The Q-factors of the two resonances are thus estimated to be \(4.6 \times 10^4\) and \(2.6 \times 10^4\) respectively. The highest measured Q-factor in the visible wavelengths is nearly half of that in the telecom wavelengths, likely due to several reasons: (1) resonant modes with higher Q-factors may exist outside our laser range; (2) the much higher mode density at visible wavelengths gives an additional loss channel into higher-order low-Q modes; (3) surface roughness becomes more comparable to the optical wavelengths, inducing more scattering loss.
2.4 Nonlinear Optical Performance in Fabricated Devices

To test the nonlinear optical properties of the fabricated single crystal LN microdisk resonator shown in Figure 2.3(a), we tuned a telecom wavelength pump laser into one of its resonances near 1546 nm and monitored the spectrum of the
transmitted light from the output port of the fiber taper using a spectrometer (HORIBA iHR550). The spectrometer data clearly show the presence of SHG signal ~ 773 nm wavelength [Figure 2.4(a)], verifying the frequency doubling process inside the LN microdisk. To further confirm this, we monitored the scattered optical signal using a silicon CCD camera (SUMIX-M81M, black and white, sensitive to wavelength < 1 µm) mounted on top of the pumped LN microdisk. Inset of Figure 2.4(a) shows the camera image, indicating the frequency doubled light traveling around the microdisk perimeter. Finally, in order to investigate the power dependence of the SHG process, we replaced the CCD camera with a highly-sensitive visible detector (HAMAMATSU, H10721-01) and measured SHG power scattered vertically for different input telecom laser power levels. The results [Figure 2.4(b)] show quadratic relationship between the output and input powers, expected from the second order nonlinear process (1). Inset of Figure 2.4(b) shows the linear fit of double-log power dependence, with a fitted slope of 1.99 ± 0.02. We calibrated the actual power received by the detector using an attenuated HeNe laser (Melles Griot, 632.8 nm) and a power meter (Newport Model 1928-C). The calculated detector sensitivity was around 40.0 A/W. Considering the detector quantum efficiencies at different wavelengths, given in
its datasheet, the detector sensitivity at 773 nm was estimated to be 1.14 A/W. The input power was calculated by assuming fiber taper losses were evenly distributed on the input and output port. When 1.83 mW of input power was coupled into the resonator, the collected SHG signal was 8.34 nA, which corresponds to 7.30 nW. Assume the SHG light was distributed uniformly in all directions, 2% of the generated light could be collected from the objective lens (NA = 0.28). Indeed the SHG light is likely coupled to higher order microdisk modes (discussed next) and the emission will be stronger in the plane of the device, which means our estimation likely gives a lower bound of the generation efficiency. The internal second harmonic intensity is thus calculated to be at least 0.365 µW. From this result, we extract a conversion efficiency of $1.99 \times 10^{-4}$ (-37 dB). When normalized by input power, the device gives a conversion efficiency of 0.109 W$^{-1}$. The normalized conversion efficiency is higher than that reported in (33) since in that case SHG signal is collected from the same fiber taper and the collection efficiency is usually low for SHG wavelengths. Our result is also comparable or better compared with SHG demonstrated in microring or microdisk resonators of similar sizes in other materials ($2.6 \times 10^{-4}$ W$^{-1}$ in GaN (20), $1.0 \times 10^{-3}$ W$^{-1}$ in SiN (48) and $5 \times 10^{-2}$ W$^{-1}$ in GaAs (49)). In real applications, it is more efficient to collect SHG signal
from a bus waveguide and then couple the light into a single mode fiber using either butt coupling (48) or grating coupler (20).

Figure 2.4 Second harmonic generation. (a) Spectrometer data showing SHG peak at 773 nm when pumped at 1546 nm, indicating frequency doubling. Inset: black-and-white CCD camera image revealing scattered SHG light travelling around the LN microdisk perimeter. The fiber taper is positioned at the relatively dimmed area, indicated by the arrow. (b) Input-output power dependence and its quadratic fit. Inset shows the linear fit of double-log curve with a slope of 1.99 ± 0.02.

Owing to the wealth of optical modes available in the 770 nm wavelength range [Figure 2.3(a)], SHG could be observed in most high-Q telecom resonances. Indeed, we note that different phase matching conditions apply to different combinations of the fundamental/second harmonic polarizations, offering more than one phase matching possibilities. For the TE/TM case, since the nonlinear
susceptibility is symmetric in plane ($\chi^{(2)}_{xxz} = \chi^{(2)}_{yyz}$), the usual phase matching condition applies: the azimuthal mode number of the second harmonic ($m_2$) equals twice that of the fundamental wavelength ($m_1$), i.e. $m_2 = 2m_1$. For the TE/TE case, on the other hand, nonlinear susceptibility is non-zero only in one direction ($\chi^{(2)}_{yyy} \neq 0, \chi^{(2)}_{xxx} = 0$), giving additional phase matching conditions, i.e. $m_2 = 2m_1 \pm 1, m_2 = 2m_1 \pm 3$. Rigorous derivation of these arguments can be derived from the methods introduced in Ref. (50). Our simulation (data not shown) shows that these phase-matched modes are usually closely packed near SHG wavelengths, so that there is almost always finite mode overlap between fundamental and second harmonic wavelengths. Importantly, we believe that our current devices are not perfectly mode-matched due to a limited parameter space available for dispersion engineering in microdisk resonators. A more efficient SHG could be achieved in a dispersion engineered ring resonator for example (51), which will be considered in our future work.

2.5 Power Handling and Time-Domain Measurements

Due to the high quality factors in our devices, optical bistability effects (52) were observed at elevated input powers, as is shown in Figure 2.5(a). As the pump laser
wavelength is tuned across the LN microdisk cavity resonance from blue to red at sufficiently high input power, increased intracavity energy results in a shift in local refractive index. The cavity stays in resonance with the laser wavelength until it abruptly jumps out of resonance when the red detuning becomes too large. Optical bistability in our devices also leads to a clear hysteresis when setting the laser at different red-detuned frequencies and monitoring input-output power relations for both upwards and downwards power sweeps [Figure 2.5(b)]. A fiber coupled EO modulator (Lucent 2623NA) was installed into the input port of the fiber taper, before the inline fiber polarizer, to modulate the input power in real time. The in-coupled powers in Figure 2.5(a-b) have accounted for losses from the tapered fiber transmission (~ 5 dB) and the EO modulator (~ 5 dB), as well as an on-resonance coupling efficiency near 50% (estimated from the resonance transmission depth). The onset of the power hysteresis loop is seen at detuning larger than 80 pm. The observed optical bistability can originate from a number of nonlinear refractive index tuning mechanisms (53, 54), including absorption-induced thermal effects (slow) and third-order optical nonlinearities (fast). To elucidate its origin, we further investigated the bistable response through time-domain measurements [Figure 2.5(c-e)]. Here, the pump laser wavelength was red-
detuned from the cavity resonance and the input power is modulated with a sinusoidal signal. The output signal from a fast InGaAs photodetector (New Focus Model 1811 125-MHz photoreceiver) was monitored using an oscilloscope. For low frequency modulation (5 kHz), distortion is clearly present, and is attributed to output signal being switched from one bistable state to the other. Distortion (bistable behavior) starts to abate as the modulation frequency is increased, and completely disappears at 100 kHz. Based on these results, we estimate a time constant of the observed bi-stable behavior to be on the order of ~ 10 µs. This indicates that the origin of bistability is likely thermal, induced by absorption from defects inside LN and/or various surface states resulting from our fabrication process. These effects may be eliminated by further optimization of both our fabrication procedure and the LNOI platform itself.
Figure 2.5 Optical bistability and its time-domain response. (a) Transmission spectra of a LN microdisk resonance at different in-coupled power levels, showing red-shift of resonant frequency and optical bistability at elevated input power. The baselines of each spectrum are normalized by input power. (b) Power hysteresis curves at different red-detuned laser wavelengths (legend indicates detuning). The arrows show the directions to which the laser power is swept. (c-e) Time-domain power responses showing both input optical signals (red) and output detector signals (blue) at different modulation frequencies: (c) 5 kHz, (d) 20 kHz and (e) 100 kHz.
Chapter 3

Phase Matched Second Harmonic

Generation in Nanostructured Lithium Niobate Waveguides

$\chi^{(2)}$ wavelength conversion processes (e.g. SHG, SFG, DFG and SPDC) not only are crucial for accessing new spectral ranges in classical optics (55-57), but also act as key resources for non-classical light generation in quantum information processing (58-60). Efficient energy transfer between waves that participate in this three-wave mixing process can only occur when the phase velocity of the nonlinear polarization and the generated field at $\omega_3$ are matched. In this case, the frequency conversion process is said to be phase-matched. This is equivalent to stating that momentum is preserved in the process, or $k_3 = k_1 \pm k_2$. Traditional phase-matching takes advantage of birefringent nonlinear-optical crystals which
allow adjustment of a wave’s refractive index by judiciously choosing the propagation angle and polarization with respect to the ordinary and extraordinary crystalline axis (assuming a uniaxial crystal symmetry for simplicity). Due to material dispersion, this equality tends to be satisfied only for a particular combination of wavelengths for a given temperature \( (1) \).

In traditional wavelength conversion devices made of LN or other ferroelectric materials, the phase matching problem is usually solved by a periodic reversal of the ferroelectric domain orientation via the application of a large electric field (electric-field poling). This so-called quasi-phase matching (QPM) technique flips the phase of the nonlinear polarization in a periodic fashion, resulting in a monotonic energy transfer between the pump and signal wavelengths \((61)\). The required poling period is determined by twice the coherence length \( L_c = \pi/\Delta k \), with the phase mismatch \( \Delta k = k_3 - k_1 \pm k_2 \). Equivalently, this periodic domain reversal adds a grating k-vector \( K_{QPM} \) to compensate for the momentum mismatch \( \Delta k \), resulting in \( k_3 = k_1 \pm k_2 + K_{QPM} \).

Figure 3.1 shows a comparison of SHG signal amplitude in the cases of perfect phase matching, QPM and phase mismatch. Owing to the flexibility in operation wavelengths, conventional QPM devices based on periodically poled lithium
niobate (PPLN) have been widely used for a range of different applications. However, these PPLN devices are usually built with weakly confined waveguides. The low index contrast ($\Delta n \sim 0.02$) between waveguide core and cladding results in large device dimensions (~ cm long and ~ 10 µm wide) and large bending radii (mm scale) (62), preventing dense integration and resonator configuration.

![Figure 3.1](image)

Figure 3.1 Comparison of second harmonic generated signal amplitude in the cases of perfect phase matching, quasi-phase matching, and phase mismatch (1).

In this chapter, we demonstrate phase matched SHG in thin-film LN nanowaveguides defined by direct dry etching, with normalized conversion efficiencies as high as 41% W$^{-1}$cm$^2$. This is enabled by the ability to precisely engineer the dispersion properties and device dimensions of the LN
nanowaveguides by the top-down fabrication method we use. To achieve the phase matching condition in our devices, we theoretically propose and experimentally demonstrate two distinct schemes: (1) modal phase matching between 1st and 3rd order TE modes in waveguides of fixed width; (2) QPM in periodically grooved lithium niobate (PGLN) waveguides. We show that both methods feature unique advantages and are promising for future integrated nonlinear wavelength conversion systems.

Figure 3.2 (a) Cross-section schematic of the x-cut LNOI waveguide, where the coordinates are aligned with the LN crystal directions. (b) Mode effective indices as a function of waveguide top width at both pump and SH wavelengths. (c) $E_z$ components of the corresponding modes at both wavelengths.
3.1 Lithium Niobate Nanowaveguide Dispersion Relations

Figure 3.2(a) displays the cross-section schematic of a typical x-cut thin-film LNOI waveguide cladded in silica. The geometric parameters of such a waveguide include top width $w_t$, bottom width $w_b$, thickness $t$, and sidewall angle $\theta$ (introduced by the dry etching process). The coordinates in Figure 3.2(a) are aligned with the crystalline directions of LN, where $z$ is the extraordinary axis. This waveguide geometry supports both TE like and TM like modes. However, here we are interested in the TE modes only in order to access the largest nonlinear coefficient $d_{33}$. Figure 3.2(b) shows the dependence of effective mode indices ($n_{\text{eff}}$) of both fundamental mode at pump wavelength (~ 1550 nm) as well as fundamental and higher order modes at second harmonic (SH) wavelength (~ 775 nm) as a function of waveguide top width ($w_t$). Representative modal profiles of $E_z$ components at both wavelengths are displayed in Figure 3.2(c). The results were obtained using a Finite Difference Eigenmode solver (MODE Solutions, Lumerical). In the simulation, we use $t = 400$ nm and $\theta = 40^\circ$, which are taken from actual device dimensions. To achieve phase matching, $n_{\text{eff}}$ at both wavelengths need to be equal. In our waveguides this cannot be achieved for fundamental TE
modes at pump and SH wavelengths, due to both material and waveguide dispersions. In the following sections, we show two methods to address the phase mismatch issue based on the waveguide dispersion displayed in Figure 3.2(b).

3.2 Modal Phase Matching Between 1\textsuperscript{st} and 3\textsuperscript{rd} order TE modes

To achieve modal phase matching, higher order modes at SH wavelength can be used to bring down the \( n_{\text{eff}} \) to match with that of the fundamental mode at pump wavelength. Note that conversion between TE\(_1\) mode at fundamental wavelength and TE\(_2\) mode at SH wavelength is prohibited by symmetry. Therefore the lowest possible phase-matching mode at SH wavelength is TE\(_3\). In fact, Figure 3.2(b) suggests that phase matching is readily achievable with \( \omega_t \sim 580 \) nm.

According to coupled-mode theory and assuming the low-conversion limit (63), the SHG conversion efficiency in a waveguide can be solved as:

\[
\gamma = g^2 L^2 P_0 \left( \frac{\sin(\delta L/2)}{\delta L/2} \right)^2 \tag{3.1}
\]

where \( \delta = \beta_{2\omega} - 2\beta_\omega \) is the phase mismatch, \( \beta_q \) is the propagation constant, \( q \in [\omega, 2\omega] \) represents the corresponding optical frequencies, \( L \) is the waveguide length, \( P_0 \) is the input optical power. Here, the overlap factor is:
\[ g = \frac{\omega}{4} \iint (E^{2\omega}(x,z))^* d_{33}(x,z)(E^\omega(x,z))^2 \, dx \, dz \]  

where \( E(x,z) \) is the normalized electric field distribution in the waveguide cross section, and \( d_{33} \) is the diagonal nonlinear coefficient of LN.

Note that, Equation [3.1] has the same form as SHG in a bulk nonlinear crystal. Considering the modal phase matching case (\( \delta = 0 \)), the normalized (by length and input power) conversion efficiency \( \eta \) is simply \( g^2 \).

Using practical waveguide parameters (\( w_t = 600 \) nm, \( w_b = 1270 \) nm, \( t = 400 \) nm), and mode profiles for fundamental pump and third order SH mode, we obtain a nonlinear overlap factor \( g = 0.774 \, \text{W}^{-1/2}\text{cm}^{-1} \), which corresponds to a normalized conversion efficiency \( \eta = 60\% \, \text{W}^{-1}\text{cm}^2 \). Taking into account the 3.0 dB/cm waveguide propagation measured in Section 3.5, a maximum conversion efficiency of 31\% W\(^{-1}\) can be achieved with a 20 mm long waveguide.

3.3 Periodically-Grooved Lithium Niobate (PGLN) Waveguides

Here, we demonstrate an alternative QPM technique to conventional PPLN devices, by utilizing a periodically-grooved structure to achieve QPM between fundamental TE modes at both pump and SH wavelengths. By introducing periodic modulation of the waveguide width, as shown in Figure 3.3(a), with a
period $\Lambda$, an additional momentum “kick” $\Delta k = 2\pi/\Lambda$ could be applied to the propagating electromagnetic wave, compensating for the phase mismatch $\delta$ (64).

Figure 3.3 (a) 3D cartoon of the proposed PGLN structure. (b) Simulated SHG efficiencies versus propagation length for a PGLN waveguide with a groove depth of 80 nm, in comparison with a uniform LN waveguide. (c) Calculated loss due to the leaky Bloch modes in PGLN at both fundamental and second harmonic wavelengths, as a function of groove depth. (d) Calculated normalized conversion efficiency as a function of groove depth. (e) SHG efficiency dependence on groove depth and propagation length. Inset: Enlarged view for the parameter space in vicinity to our experimental operation point (red circle). The dashed line indicates the optimal groove depth for each propagation length.
To analytically solve the nonlinear coupled-mode equations in this case, we consider the PGLN waveguide as a perturbation from the uniform waveguide case (described in Section 3.2), with both the linear permittivity \( \varepsilon \) and nonlinear coefficient \( d_{33} \) periodically modulated along the propagation direction (y-axis). This is justified since the effective index perturbations corresponding to 80 nm groove depth are within 2.3% and 0.7% for fundamental and SH wavelengths, respectively. We can therefore expand both \( \Delta \varepsilon \) (permittivity difference between waveguide and environment) and \( d_{33} \) as Fourier series in y:

\[
\Delta \varepsilon(x, y, z) = \sum_m \Delta \varepsilon_m(x, z) \exp(-jm\Delta ky)
\]

\[
d_{33}(x, y, z) = \sum_m d_{33}^{(m)}(x, z) \exp(-jm\Delta ky)
\]

In this case, we have the new overlap factor \( g' \):

\[
g' = g_{NL}^{(1)}(J_0(\varphi_L) + J_2(\varphi_L)) - g_{NL}^{(0)}J_1(\varphi_L)
\]

with

\[
g_{NL}^{(m)} = \frac{2\omega}{4} \iint (E^{2\omega}(x,z))^* d_{33}^{(m)}(x,z)(E^{\omega}(x,z))^2 \, dx \, dz
\]

\[
g_{L}^{\omega} = \frac{\omega\varepsilon_0}{4} \iint \Delta \varepsilon_1^{\omega}(x,z)|E^{\omega}(x,z)|^2 \, dx \, dz
\]

\[
g_{L}^{2\omega} = \frac{2\omega\varepsilon_0}{4} \iint \Delta \varepsilon_1^{2\omega}(x,z)|E^{2\omega}(x,z)|^2 \, dx \, dz
\]
\[ \varphi_L = \frac{2(g_L^2\omega - 2g_L\omega)}{\Delta k} \]  

[3.9]

Now, the phase mismatch term becomes \( \delta' = \beta^2\omega - 2\beta\omega - 2\pi/A \), and \( J_p \) denotes the \( p \)-th order Bessel function (only first three terms with slow spatial variation are taken). Detailed derivations can be found in Ref. (65).

The new overlap factor \( g' \) consists of two terms. The first term results from the periodically varying nonlinear coefficient \( d_{33}^{(1)} \), which is the same effect as in PPLN. The second term originates from the constant nonlinear coefficient \( d_{33}^{(0)} \) and periodically modulated dielectric constant \( \Delta \varepsilon_1 \), or grating effect.

For a typical PGLN waveguide (\( w_l = 670 \) nm, \( w_b = 1300 \) nm, \( t = 400 \) nm) with a periodic groove depth of 80 nm, the nonlinear coupling coefficient is calculated to be \( g' = 0.345 \) W\(^{-1/2}\)cm\(^{-1}\), which corresponds to a normalized conversion efficiency \( \eta \) of 12.1% W\(^{-1}\)cm\(^2\). The total conversion efficiency \( \eta \) in the low-conversion limit is plotted as a function of propagation length in Figure 3.3(b), in comparison with a uniform waveguide without periodic grooves. Here we also take into consideration the oscillating phase-mismatched optical fields, which could be obtained simply by replacing the overlap factor in Equation [3.5] with \( G' = g' + \)
\[ g_{NL}^{(0)} J_0(\varphi_L) \exp(j\Delta ky) \]. The net conversion efficiency features a quadratically increasing envelope with local oscillations.

In a realistic scenario, PGLN waveguide loss should be taken into account. There are two main loss channels in our devices: intrinsic loss due to the leaky nature of the Bloch modes supported by our waveguide (66), and roughness-induced scattering loss due to fabrication imperfections. We calculated the intrinsic loss component using the Finite Difference Time Domain method (FDTD Solutions, Lumercial) by modeling one unit cell of the corrugated waveguide and applying periodic boundary condition. The results are summarized in Figure 3.3(c). As expected, both modes are lossier for deeper grooves, which limits the propagation lengths of the modes and reduces the overall conversion efficiency. Furthermore, the second harmonic mode is a lot more sensitive to the corrugation since the mode has a larger overlap with it (see Figure 3.2(c)). On the other hand, larger groove depth ensures stronger nonlinear overlap between the two modes, and can result in larger conversion efficiency [Figure 3.3(d)]. In order to model this trade-off between the competing effects from the grooves, we introduce loss terms in the nonlinear coupled-mode equations. In addition to the intrinsic loss, we have also
included 3.0 dB/cm fabrication induced scattering loss that we experimentally obtained from our devices (Section 3.5).

Assuming that the pump is not depleted by the generated SH, and that the groove period exactly compensates for the initial phase mismatch, or $\delta' = 0$, the new conversion efficiency including the waveguide loss can be expressed as:

$$\gamma' = g'^2 P_0 \left( \frac{\exp(-2\alpha_\omega L) - \exp(-\alpha_{2\omega} L)}{2\alpha_\omega - \alpha_{2\omega}} \right)^2$$

[3.10]

where $\alpha_\omega$, $\alpha_{2\omega}$ are the loss coefficients of the PGLN waveguide for pump and SH wavelengths respectively, including both scattering loss and Bloch loss.

Figure 3.3(e) shows the theoretical result of conversion efficiency as a function of groove depth and waveguide length, with losses taken into account. The global peak in our current device configuration is present for a groove depth $\sim 22$ nm and a waveguide length $\sim 11$ mm, which correspond to a maximum conversion efficiency of 0.16% W$^{-1}$. Note this global maximum efficiency is still limited by the intrinsic propagation loss in our waveguides. For example, if the roughness-induced loss could be reduced to 0.25 dB/cm, a groove depth of 8 nm and a total length of 11 cm could yield a 2.3% W$^{-1}$ overall conversion efficiency. In our proof-of-concept experiments, we focus on shorter waveguides, with a length of 500 µm,
which results in an optimal groove depth of ~ 80 nm. In this case, the total waveguide propagation losses are 13.5 dB/cm at pump wavelength and 63.5 dB/cm at SH wavelength, resulting in an overall conversion efficiency of 0.012% $W^{-1}$ and a normalized conversion efficiency of 4.6% $W^{-1}cm^{-2}$. Although PGLN waveguide yields lower conversion efficiencies than uniform waveguides, it achieves wavelength conversion between fundamental optical modes at both wavelengths, which is beneficial in many applications.

3.4 Fabrication of Uniform and Periodically Grooved Waveguides

Starting from an x-cut LNOI substrate (400 nm thick, NANOLN), uniform and PGLN waveguides were fabricated using a process modified from that used in Chapter 2, except that an amorphous silicon (a-Si) etching mask is used instead of resist. An 800 nm thick a-Si layer was first deposited on the substrate via plasma-enhanced chemical vapor deposition (PECVD). The a-Si layer was then patterned with a combination of EBL and RIE, and used as a hard mask for the subsequent LN dry etching in Ar+ plasma. After removing leftover silicon mask in KOH (80 °C), the waveguides were cladded in silica (3 μm thick) using PECVD. Waveguide facets were diced and polished to ensure high coupling efficiency. Figure 3.4
shows the SEM images of uniform LN and PGLN waveguides without cladding. The bending sections in the waveguides are used to prevent the output fiber from collecting light directly from the input fiber. It can be seen that our fabrication process is well capable of delivering designed structures, while maintaining minimum surface roughness and manageable scattering loss.

Figure 3.4 Representative SEM images of the fabricated devices. (a) An array of LN waveguides with slightly different widths. (b) A typical uniform LN waveguide with fixed width. (c) A typical PGLN waveguide with a spatial modulation period of 2.77 µm and a groove depth of 80 nm.
3.5 Second Harmonic Generation Measurement Results

We measured the SHG response of both modal phase-matched LN and PGLN waveguides using the setup shown in Fig. 4(a). Tunable telecom lasers (TLS, Santec TSL-510, 1480 – 1680 nm) were used as light sources at pump wavelength. A fiber polarization controller (FPC) was used to ensure TE mode input before end-fire coupling into the device under test (DUT). The SHG signals with TM polarized input are usually orders of magnitude lower than TE input due to misaligned crystal relevant axes. Light is coupled into and out of the waveguide facets using tapered lensed fibers. The output light is sent to either an InGaAs photodetector (IGA PD), or a silicon avalanche photodetector (Si APD), to monitor the linearly transmitted telecom light and the SHG signal respectively.

Figure 3.5(b) shows the linear transmission spectrum (in absolute unit) of a typical uniform waveguide at telecom wavelengths. It indicates a total fiber-to-fiber loss ~ 10.2 dB, resulting from a combination of facet coupling loss and waveguide propagation loss. The fringe pattern seen in Figure 3.5(b) is a result of Fabry-Perot interference between the two polished facets of the waveguide. From the contrast
between maximum and minimum of the transmission fringes, we can extract the
waveguide propagation loss using the following equation (67):

\[ \alpha = \frac{4.34}{L} (\ln R - \ln \tilde{R}) \]  

[3.11]

with \( \tilde{R} = \frac{1}{K} (1 - \sqrt{1 - K^2}) \), \( K = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \). Here \( I_{\text{max}} \) and \( I_{\text{min}} \) are the maximum and
minimum intensities of the transmitted light. \( R \) is the reflection coefficient at the
waveguide facet, which is calculated to be 0.147 using FDTD. \( L \) is the waveguide
length, which is 3 mm in this case (including coupling and bending sections with
phase-mismatched widths). The waveguide propagation loss is thus calculated to
be 3.0 ± 0.2 dB/cm. This value represents the best propagation loss reported in thin-
film LN waveguides with sub-wavelength light confinement (\( A_{\text{eff}} = 0.52 \ \mu m^2 \) at
telecom wavelength), and is consistent with the quality factor achieved in our
previous microdisk resonators (Chapter 2). Using the calculated propagation loss
and the measured transmission, we are able to estimate the fiber-to-waveguide
coupling efficiency. By characterizing many waveguides on the same chip used in
the following experiments, an average coupling efficiency of 33% on each facet is
extracted (4.8 dB/facet). This coupling loss can be reduced to < 0.5 dB with
adiabatic mode transition using tapered fibers (68).
Figure 3.5(c-d) shows the measured SHG efficiencies as a function of SH wavelength for both modal phase matched LN waveguides (1 mm long) and quasi-phase matched PGLN waveguides (0.5 mm long). These values have accounted for the APD spectral quantum efficiency and the facet coupling loss, and are normalized by the pump laser power. For each scheme, a set of waveguides with slightly different widths are characterized. SHG peaks can be clearly observed at the corresponding (quasi-) phase matching wavelengths. The SHG peak wavelength changes with increasing waveguide width in both cases, but in opposite directions, which agrees with theoretical prediction. For the modal phase-matched waveguide with 630 nm top width, we measured SHG power of 1.38 pW at 18.3 μW pump power, which corresponds to a normalized conversion efficiency of 41% W⁻¹cm². Similarly, for PGLN with a top width of 670 nm, the measured output power is 0.334 pW at 44.3 μW pump power, or 6.8% W⁻¹cm². The dotted curves in the insets of Figure 3.5(c-d) show the theoretical SHG bandwidths and peak intensities calculated using the methods described in Section 3.2 and 3.3. In both schemes, the measured bandwidths match the theoretical predictions well, while the peak efficiencies are slightly lower than theory, indicating possible inhomogeneity in the waveguide dimensions throughout the chip.
To confirm that the generated SH light couple to third-order and fundamental mode in the case of modal and groove-assisted phase matching, respectively, we used a high NA (0.8) lens and a black & white visible CCD camera to monitor the scattered SH light at the output facet [Figure 3.5(e-f)]. In the case of uniform LN waveguide [Figure 3.5(e)], the SHG signal clearly radiates in three lobes, indicating SH light being generated in the 3rd order transverse mode. In comparison, the SH light generated in the PGLN waveguide [Figure 3.5(f)] is in the fundamental mode and radiates only in one lobe.
Figure 3.5 (a) Schematic of the measurement setup. (b) Transmission spectrum of a typical uniform LN waveguide. Inset: zoom-in view of the wavelength range used for calculating propagation loss. (c-d) Conversion efficiency versus SHG wavelength for (c) uniform LN waveguides and (d) PGLN waveguides with different waveguide widths (measured at the waveguide top). Insets: comparison between experimental (solid) and theoretical (dotted) SHG efficiencies and bandwidths. (e-f) CCD camera images of the scattered SHG light at the output facets of a uniform LN waveguide (e) and a PGLN waveguide (f), indicating the corresponding output optical modes.
Chapter 4

Lithium Niobate Metasurfaces for Linear and Nonlinear Optics

The phase-matching condition is a key aspect in nonlinear wavelength conversion processes, which requires the momenta of the photons involved in the processes to be conserved. Conventionally in bulk nonlinear crystals, as we have discussed in Chapter 3, phase matching can be achieved by using crystal birefringence, by controlling the angle of intersection between optical beams, or by using periodically poled nonlinear crystals (i.e. quasi-phase matching) (1).

Nonlinear effects can be significantly enhanced inside nanophotonic waveguides with tight light confinement (21, 22, 39, 49, 51, 69-71). Nanophotonics also provides alternative tools to achieve phase-matching, e.g. dispersion engineered structures (22, 51, 69, 70), photonic crystal waveguides (21, 71), and anisotropic crystal orientations (49), in addition to more traditional approaches like periodic poling.
Nevertheless, the nonlinear phase-matching condition still needs to be satisfied between co-propagating light waves. On the other hand, strict phase-matching is not required in resonant structures with (sub-) wavelength-scale modal volumes, where the conversion efficiency relies on modal overlap between fundamental and higher harmonic modes (23, 72-84). For example, doubly resonant photonic cavities with high quality factors have been proposed to achieve highly-efficient wavelength conversion, while their narrow bandwidth remains the major challenge in experiments (23, 72-74). Plasmonic nano-antennas has also been used to achieve enhanced optical nonlinearities (75-80). Simultaneous field enhancement at both the fundamental and harmonic wavelengths and spatial overlap between the modes at these wavelengths allow for even higher conversion efficiency (81, 82). More recently, dielectric antennas have emerged as a promising alternative over plasmonic ones due to lower optical loss and high damage threshold. Silicon Mie resonators supporting magnetic dipole resonance (83) and Fano resonance (84) have been utilized for enhanced third harmonic generation. However, these devices suffer from low conversion efficiency due to limited light-matter interaction distance.
In this Chapter, we leverage the high nonlinear susceptibility of LN and the strong capability of metasurfaces in controlling the modal indices of waveguide modes, to efficiently manipulate the waveguiding behavior in both linear and nonlinear optics. We first show that the unidirectional effective wavevector provided by gradient metasurface can be used to convert between waveguide optical modes with high efficiency and transmission. We further utilize this hybrid metasurface/waveguide structure to demonstrate a phase-matching-free SHG process. The gradient metasurface enables a one-way transfer of optical power from the pump to the SH signal, which represents a new scheme of nonlinear phase matching that is fundamentally different from conventional methods. In this new scheme, the efficiency of nonlinear optical generation is no longer sensitively dependent on the pump frequency and device geometry. We demonstrate efficient, broadband, and robust SHG, where the SH signal monotonically increases over a distance of many coherent lengths.
Figure 4.1 (a) Schematic of an integrated nonlinear photonic device. Here three antenna arrays are shown. For linear mode converter only one antenna array is used. (b) Schematic of the device cross-section.

4.1 Linear Waveguide Mode Converter

The gradient metasurface in our devices consists of a phased array of dielectric antennas patterned on the top surface of a nonlinear optical waveguide (Figure 4.1). We utilize the dipolar Mie resonances in dielectric nano-rod antennas of different lengths to introduce different values of phase of the scattered light waves. Collectively, the phased antenna array introduces a unidirectional phase gradient \( \frac{d\Phi}{dz} \), which is equivalent to a unidirectional effective wavevector \( \Delta k \), along the waveguide. Here, \( d\Phi \) is the difference in phase response between adjacent nano-antennas that are separated by a subwavelength distance of \( dz \). The unidirectional effective wavevector \( \Delta k \) enables directional coupling of waveguide modes. That is, when an incident waveguide mode propagates against \( \Delta k \), the modal index...
decreases, which corresponds to coupling into higher-order waveguide modes; however, the inverse process in which optical power is coupled from the higher order modes to the lower-order ones will be prohibited due to the lack of a phase-matching mechanism.

In this section, we utilize this gradient metasurface to demonstrate an efficient TE-TM mode converter in integrated LN waveguides. We choose amorphous silicon (a-Si) as the material for the dielectric phased array antennas. A-Si has a refractive index (~3.5 at telecom wavelength) significantly higher than LN (~2.1). Therefore, there will be a strong interaction between waveguide modes in LN and Mie resonance modes in the a-Si nano-antennas. The use of dielectric antennas instead of plasmonic antennas minimizes the absorption losses.

Figure 4.2 shows the design and simulation results for several different mode converters at telecom wavelength. We design highly efficient mode conversion between fundamental TE\textsubscript{11} mode and TM\textsubscript{11}, TE\textsubscript{21}, TE\textsubscript{31} modes based on the dielectric gradient metasurface configuration shown in Figure 4.1. Here the lengths of the three mode converters are 20.3 \( \mu \)m, 5.4 \( \mu \)m and 8.4 \( \mu \)m respectively. The calculated transmission efficiencies of the three devices from top to bottom averaged over \( \lambda = 1,480 \sim 1,580 \) nm are 94.4\%, 92.9\% and 82.9\%, respectively. The
total number of effective wavevectors $k_{\text{eff}} = d\Phi/dx$ imparted to the guided wave by the gradient metasurfaces during the mode conversion process is 48.2, 2.6 and 4.3, respectively.

Figure 4.2 Simulated waveguide mode converter performance at $\lambda = 1.55 \mu$m. First column: Schematics of three waveguide mode converters. Second and fourth columns: Waveguide modes at the input and output ports of the devices, respectively. The polarization of the electric field component of the modes is indicated by arrows. Third column: Mode evolutions as light propagates from left to right.
Figure 4.3(a) shows the fabrication procedure for the gradient metasurface on LN waveguides. X-cut LNOI with a 400 nm thick device layer bonded on top of a 2µm thick silica buffer layer, were used for device fabrication. LN ridge waveguides were first defined using EBL (with HSQ resist) and the same Ar⁺ plasma etching technique as is described in Chapter 2. The LN ridge waveguides have a top width of 1 µm and a height of 300 nm, leaving a 100 nm thick LN slab underneath. The residue HSQ resist was removed in BOE. A 145 nm thick p-doped a-Si layer was then deposited on top of the entire sample surface using PECVD. A second HSQ resist layer (~ 300 nm) was then patterned on the a-Si surface using EBL. RIE was performed to transfer the second HSQ pattern into the a-Si layer, defining the antenna arrays. The antennas have a width of 105 nm and a rotation angle of 60°. The gap between adjacent antennas is 80 nm. Finally a second BOE etch was used to remove the residue HSQ resist, before the device is cladded in SiO₂ by PECVD. Figure 4.3(b-d) presents SEM images of a fabricated telecom TE₁₁-to-TM₁₁ mode converter according to the design shown in Figure 4.2.
Figure 4.3 (a) Schematic showing the procedure of device fabrication. (b) SEM image of a fabricated TE$_{11}$-to-TM$_{11}$ mode converter (before applying an overcoat of SiO$_2$). (c-d) Zoom-in views of (b).

The devices were characterized by launching light from a tunable laser source ($\lambda = 1,480 \sim 1,580$ nm) into the TE$_{11}$-to-TM$_{11}$ mode converters as well as into bare waveguides with the same length and cross-sectional dimensions as the mode converters. We measured mode conversion with high purity and broad bandwidth on these devices. Figure 4.4 presents the measured spectra of the TE-and TM-
polarized components of a TE_{11}-to-TM_{11} mode converter and of a bare waveguide, respectively. The measured TE and TM components must be contributed by the TE_{11} and TM_{11} waveguide modes, respectively, because the LN waveguides are designed to only support the fundamental TE and TM modes. Thus, the spectra in Figure 4.4(b) indicate that the TE_{11} mode is coupled into a bare waveguide and propagates along the entire length of the waveguide (i.e. 2 mm), while maintaining high modal purity (> 99%). Likewise, Figure 4.4(a) indicates that when the dielectric gradient metasurface is patterned on the waveguides, the TE_{11} mode launched into the device is converted into a high-purity TM_{11} mode (purity ~ 96% averaged over \( \lambda = 1,480 \sim 1,580 \) nm).

In addition, we observed no significant difference in the transmitted power between the mode converters and the bare waveguides, implying that the dielectric metasurfaces introduce negligible insertion losses. For example, the measured transmitted optical signal from an InGaAs photodetector for the TE_{11}-to-TM_{11} mode converters averaged over six devices is 1.09 ± 0.14 V, while the measured transmitted optical signal of 10 bare waveguides is 1.07 ± 0.19 V.
Figure 4.4 (a) Measured TE- and TM-polarized components at the output port of a TE$_{11}$-to-TM$_{11}$ mode converter, showing that the device operates over a broad wavelength range. (b) Measured TE- and TM components at the output port of a bare waveguide that has the same geometries as the mode converter in (a).
4.2 Phase-Matching-Free Second Harmonic Generation

Next, we use the gradient metasurface discussed in Section 4.1 to demonstrate phase-matching-free SHG. Figure 4.5 shows the working principle of the metasurface-based nonlinear photonic device. In the region of the nonlinear waveguide patterned with the metasurface structure, once optical power couples from the fundamental mode at the pump frequency, $\text{TE}_{11}(\omega)$, to the fundamental mode at the SH frequency, $\text{TE}_{11}(2\omega)$, it instantly starts to get converted into higher-order waveguide modes at the SH frequency, $\text{TE}_{mn}(2\omega)$ and $\text{TM}_{mn}(2\omega)$, by the gradient metasurface. The unidirectional wavevector provided by the gradient metasurface ensures that optical power cannot be coupled from higher order modes, $\text{TE}_{mn}(2\omega)$ and $\text{TM}_{mn}(2\omega)$, back to $\text{TE}_{11}(2\omega)$ (dotted arrow in Figure 4.5(c)). Furthermore, the optical power carried by $\text{TE}_{mn}(2\omega)$ and $\text{TM}_{mn}(2\omega)$ cannot be coupled back to the pump, $\text{TE}_{11}(\omega)$ (dashed arrow in Figure 4.5(c)), because the coupling coefficient between them is negligible (i.e., spatial overlap between $\text{TE}_{mn}(2\omega)/\text{TM}_{mn}(2\omega)$ and $\text{TE}_{11}(\omega)$ on the waveguide cross-section is very small). Optical power is thus retained in the SH wavelength and accumulates as a function of propagation distance. The antennas are designed to interact with guided waves
at the SH frequency, and their sizes are too small to strongly scatter the pump. In this way, asymmetric optical power transfer is realized between the pump and the SH signal (i.e., process indicated by arrow #1 is more efficient than process indicated by arrow #5) and as a result the SHG power will monotonically increase as a function of the propagation distance. The order of waveguide modes at the SH frequency, however, keeps increasing; as such, the SH signal will eventually leak out from the waveguide when the cutoff condition for waveguiding is reached. This new scheme circumvents the nonlinear phase-matching condition as long as the gradient metasurface provides an asymmetric effective wavevector to preferentially couple optical power from lower to higher-order waveguide modes at the SH frequency and does not interact with the pump.

Figure 4.5 Conceptual diagram of the proposed phase-matching-free SHG.
The devices used in this case have very similar structures as those described in Section 4.1. Here the ridge LN waveguides have a top width of 2600 nm. Each antenna array consists of 35 a-Si nano-antennas with a thickness of 75 nm, a width of 75 nm, and a range of lengths. The separation, dz, between adjacent nano-antennas is 140 nm and the phase difference, dΦ, between them is 0.5 degrees at the SH frequency (i.e. \( \lambda \sim 750 \) nm).

Figure 4.6 (a) Simulations showing that the SH signal increases monotonically as the number of phased antenna arrays increases. Without the antenna arrays, the SH signal oscillates periodically along the waveguide (black curves).
Figure 4.6 shows simulated performance of our devices in comparison to that of bare nonlinear waveguides. In a bare waveguide, the generated SH signal will be carried by the TE_{11}(2\omega) mode because of the efficient nonlinear overlap between the two fundamental waveguide modes, TE_{11}(2\omega) and TE_{11}(\omega) (i.e. integration of TE_{11}^{*}(2\omega) \times TE_{11}^{2}(\omega) over the waveguide cross-section is large). However, due to the large phase mismatch between the two modes, optical power will be frequently exchanged between them along the waveguide (with a period of twice the coherent buildup length; oscillatory black curves in Figure 4.6; thus, the SH signal can never reach a high intensity. However, when the nonlinear waveguide is patterned with the gradient metasurface structure, the SHG power monotonically increases as a function of the propagation distance, and a longer metasurface structure consisting of more sets of phased antenna arrays produces SH signals with higher intensities. For example, using just one set of phased antenna array (4.76 \mu m in length), the SHG power is increased by 7 times compared with the peak value achievable in a bare waveguide. Using six sets of phased antenna arrays with a total length of 28.6 \mu m, the SHG power can reach a value that is two orders of magnitude higher than that is achievable in a bare waveguide (Figure 4.6). Increasing the number of phased antenna arrays can further improve the
nonlinear conversion efficiency, until a point at which the generated SH signal is coupled by the gradient metasurfaces into leaky waves. Our simulations show that with the current device configuration, a monotonic increase of SHG power could be sustained for at least 10 sets of phased antenna arrays (< 50 μm in length).

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Figure 4.7 Simulations showing the evolution of the pump and the SH signal as they propagate along the waveguide. Each figure is normalized so the peak value is one.

Figure 4.7 shows the mode evolution at both the pump and SH frequencies as a function of the propagation distance in a waveguide section patterned with three
sets of phased antenna arrays. The simulation results show that the pump power is primarily carried by the fundamental waveguide mode, TE$_{11}$(ω), throughout the metasurface-patterned region, indicating a weak interaction between the pump and the metasurface structure. At the SH frequency, the x-component of the electric field has mainly a single lobe, which indicates that optical power is converted from the TE$_{11}$(ω) mode to the TE$_{11}$(2ω) mode. Furthermore, the electric field polarized along the y-axis (x-axis) at the SH frequency shows multiple lobes, and this is the result of coupling of optical power from the TE$_{11}$(2ω) mode to higher-order TM$_{mn}$(2ω) (TE$_{mn}$(2ω)) modes by the gradient metasurface.

The asymmetric coupling between the pump and the SH signal as a result of the unidirectional wavevector provided by the gradient metasurface makes the SHG process tolerant to the variations of the pump frequency and the geometry of the device. Figure 4.8 shows the simulated SHG power as a function of the pump wavelength ranging from 1500 nm to 1650 nm, for different sets of phased antenna arrays. In the case of one set of antenna array, the generated SHG power is almost a constant for different pump wavelengths. When the number of the antenna arrays is increased to six, the wavelength range within which the SHG power is above 80% of the peak value is still larger than 115 nm.
Figure 4.8 Simulations showing that the SHG power increases as the number of the phased antenna arrays increases and that the SHG process is broadband (i.e. efficient SHG is observed over a wide range of pump wavelengths).

Figure 4.9 shows that the SHG process is also robust against the variation of the device geometry. The change to the SHG power in a device with three sets of phased antenna arrays is small when the lengths of the nano-rod antennas deviate from their designed values by ±10% [Figure 4.9(a)], when the antenna arrays are offset from the center of the waveguide up to 100 nm [Figure 4.9(b)], and when the width of the LN waveguide deviates from its designed values by ±100 nm [Figure 4.9(c)]. In comparison, in conventional schemes of nonlinear phase matching, the
nonlinear wavelength conversion process is typically sensitively dependent on the optical alignment, pump wavelength, and operating temperatures (I).

Figure 4.9 Simulations showing that within the new scheme of phase-matching-free nonlinear generation the SHG process is insensitive to the variations of device geometries, including the lengths of nano-rod antennas (a), the offset of the antenna arrays from the center of the waveguide (b), and the waveguide width (c). The devices were fabricated using the same procedure as shown in Figure 4.3. Figure 4.10 shows SEM images of a representative fabricated device. The
fabricated devices were characterized using a butt-coupling setup. Telecom pump light from a continuous wave tunable laser was coupled into the LN waveguides through a tapered lensed fiber. A fiber polarization controller was used to ensure TE mode input. SH light was collected from the output waveguide facet using a second tapered lensed fiber and measured using a silicon APD. The scattered SH light was monitored by placing a visible CCD camera on top of the device, focused on the metasurface section.

Figure 4.10 SEM image of a fabricated device showing four phased antenna arrays consisting of silicon nano-rods of different lengths patterned on the top surface of a LN waveguide. Right: zoom-in view of the device (dashed frame in the left SEM image).

Figure 4.11 shows the measured SHG power from several devices that have the same LN waveguide patterned with different sets of phased antenna arrays, as well as SHG power produced by a control bare waveguide. Over the range of $\lambda_{pump}$
= 1580 – 1650 nm, the SHG process in all devices with the antenna arrays shows significant enhancement compared with the bare LN waveguide. The enhancement of the SHG process increases with increasing number of antenna arrays [Figure 4.11(a)], which is in good agreement with the simulation results shown in Figure 4.6 and Figure 4.8. To further confirm that the SHG enhancement is indeed contributed by the antenna arrays, a visible camera is used to visualize the scattered SH light from the devices. The SH signal is most likely to be scattered at the boundaries [labeled as “input” and “output” in Figure 4.11(b)] between the metasurface-patterned section of the LN waveguide and sections of bare LN waveguide due to abrupt modal index changes. At three arbitrarily chosen pump wavelengths $\lambda_{\text{pump}} = 1610, 1630, \text{and } 1650 \text{ nm}$, the scattered SH light is observed only at the output end of the antenna arrays, but not at the input end [Figure 4.11(b)], indicating that the SH signal is generated within the metasurface section. Figure 4.11(c) shows the input-output power relations for a device patterned with four phased antenna arrays. Quadratic power dependence is observed at three pump wavelengths, indicating a broadband second-order nonlinear optical process.
Figure 4.11 (a) Measured SHG power at the output port of a few devices patterned with different number of phased antenna arrays, in comparison with that of a bare LN waveguide. (b) Top-view camera images showing the scattered SH light from a device patterned with a gradient metasurface consisting of four phased antenna arrays (indicated by the white dashed frames). (c) Input-output power relations showing quadratic power dependence between the pump and the SH signal at three different pump wavelengths. (d) SHG peak (accidental phase matching) at a pump wavelength of 1660 nm, which exists in all tested devices including bare LN waveguides. (e) Optical camera image at the accidental phase-matching wavelength (1660 nm).

Note that there is a SHG peak at $\lambda_{\text{pump}} = 1660$ nm for all tested devices, including bare LN waveguides [Figure 4.11(d)]. This is the result of an accidental phase-matching between the TE$_{11}$($\omega$) mode and the TE$_{17}$(2$\omega$) mode for our LN waveguide dimensions (Figure 4.12). At this peak wavelength, scattered SH light is observed at both the input and output ends of the antenna arrays [Figure 4.11(e)], indicating
that the SH signal has already been generated before light interacts with the antenna arrays. In the case of a bare LN waveguide and at $\lambda_{\text{pump}} = 1660$ nm, as a result of the coherent accumulation of SH signal over the 1.5-mm long waveguide, the peak SH intensity is orders of magnitude stronger than the phase-mismatched SH signal at other pump wavelengths. The intensity of the broadband enhanced SH signal from the device with four phased antenna arrays is comparable to that of the phase-matching peak at $\lambda_{\text{pump}} = 1660$ nm, although the interaction length between the pump and the antenna arrays is less than 20 $\mu$m. This suggests that the SHG process assisted by the gradient metasurfaces is highly efficient.

The measured SHG spectra are not as flat as that in simulations [Figure 4.11(a)], which is likely due to two reasons: (1) After light propagates through the metasurface-patterned waveguides, there is a phase-mismatched interaction between the SH signal and the residual pump, which results in a small oscillation of the SH intensity as a function of the propagation distance. The spatial oscillation of the SH intensity and the dependence of the oscillation period on the pump wavelength lead to a variation of the SH signal measured at the output port of the LN waveguide as a function of the pump wavelength. (2) The SH signal is carried by a different combination of higher-order waveguide modes at different pump
wavelengths, which leads to different overall collection efficiencies by the tapered lensed fiber.

Figure 4.12 Simulated modal effective index vs. pump wavelength for different optical modes at both wavelengths, indicating accidental phase matching point ~ 1660 nm.
Nanophotonic Lithium Niobate

Electro-Optic Modulators

Data centers, metropolitan and long-haul data communication networks demand scalable and high-performing electro-optic modulators to convert electrical signals to modulated light waves at high speed (2, 85). For decades, LN has been the material of choice owing to its excellent properties - namely large electro-optic response, high intrinsic bandwidth, wide transparency window, exceptional signal quality and good temperature stability (2, 4, 85). Existing LN modulators however are not scalable due to the difficulty in nanostructuring LN (85). As a result, they remain bulky (~ 10 cm long), discrete, expensive, and require high-power electrical drivers (2, 85).

To address the need of a scalable and high-performance electro-optic modulator for modern data communications, tremendous efforts have been made towards a
variety of platforms that feature small footprints and high data bandwidths, including Si (86-89), InP (90-92), AlN (93), plasmonics (94), graphene (95-97) and polymers (98, 99). However, these approaches have fundamental material limitations that are hard to overcome. For example, both Si and InP modulators rely on switching mechanisms (carrier injection and quantum-confinement Stark effect, respectively) that are intrinsically nonlinear, absorptive, and sensitive to temperature fluctuation (86-92). Limitations of the other platforms include low switching efficiency (AlN) (93), high optical loss (plasmonics) (94), challenging scalability (graphene) (97) and poor long term stability (polymer) (99).

LN remains the preferred material for high-performance electro-optic modulators due to its wide bandgap (high transparency) and large second order ($\chi^{(2)}$) electro-optic coefficient (27 pm/V). In contrast to Si and InP, the $\chi^{(2)}$ process in LN changes its index of refraction linearly with an applied electrical field, at femtosecond timescale. The efficiency of this process is determined by the overlap of the optical and the electrical fields. Conventional ion-diffused LN waveguides suffer from the low refractive index contrast ($\Delta n < 0.02$) between core and cladding (Figure 1.1), resulting in large optical mode areas and bending radii (12). As a result, the photonic structures are large and the radio-frequency (RF) electrodes have to be
placed far away from the optical mode to prevent detrimental waveguide propagation loss, significantly reducing the electro-optic switching efficiency (2, 85).

Thin-film LNOI has recently emerged as a promising candidate to shrink the optical mode volume and boost the electro-optic efficiency (27). While progress has been made towards chip-scale LN devices (27, 29-31, 34, 38-40, 100-102), realization of a monolithic LN nanophotonic circuit remains challenging due to the difficulty in LN dry etching. Instead, heterogeneous integration of intact LN thin films with another easy-to-etch material has been pursued (30, 31, 40, 100). These hybrid platforms, however, suffer from low bandwidth performance and/or low optical confinement due to the addition of non-ideal deposited materials. For example, the hybrid Si/LN modulator has a bandwidth ~ 5 GHz due to the large resistance of the bonded silicon layer (30). Ridge waveguides formed by depositing passive index-matching materials (e.g. silicon nitride and chalcogenide) results in weakly confined optical modes and large bending radii (> 0.2 mm) (31, 40, 100). Furthermore, the electro-optic efficiencies in these hybrid devices are also compromised due to the reduced overlap between the electric field and the active material region (30, 31, 40, 100).
In this chapter, we demonstrate monolithically integrated LN electro-optic modulators that are orders of magnitudes smaller and more efficient than traditional bulk LN devices while preserving LN’s excellent material properties. Our compact LN platform consists of low-loss nanoscale LN waveguides, micro-ring resonators and miniaturized Mach-Zehnder interferometers (MZI), fabricated by directly shaping LN thin films into sub-wavelength structures. The efficient confinement of both optical and microwave fields at the nanoscale dramatically improves the device performances featuring a half-wave electro-optic modulation efficiency of 1.8 V\( \cdot \)cm while operating at data rates up to 40 Gbps.

5.1 Design and Fabrication of the Optical Devices and Electrical Contacts

In contrast to traditional LN modulators, our nanophotonic solution offers single-crystalline LN photonic structures with submicron optical confinement, small bending radii (< 20 µm) and low propagation loss by directly shaping single-crystalline LN into nanoscale waveguides (Figure 5.1). In our thin film LN devices, the index contrast between the LN core and the SiO\(_2\) cladding is \( \Delta n \sim 0.67 \), over an order of magnitude higher than ion-diffused LN waveguides. As is shown in
Figure 5.2, devices are fabricated from an x-cut LNOI substrate (NANOLN), here with a 700 nm LN thin film. Waveguides and resonators are first defined and etched using the same method described in Chapter 3 (Ar+ plasma etching with an 800 nm thick a-Si hard mask). Waveguide are aligned such that light propagation is perpendicular to the crystal z-axis. Here the LN etch depth is ~ 400 nm, leaving a 300 nm slab across the chip. Bottom metal electrodes (15 nm Ti/300 nm Au, deposited using an electron-beam evaporator) are formed along the MZI waveguides and racetrack straight arms using a standard PMMA/MMA double layer lift-off process. A 1.5 μm thick silica cladding layer is then deposited on top of the waveguides and electrodes using PECVD. A new PMMA resist is spun coated and written with EBL to open windows at corresponding via locations. Vias are formed by 5 min wet etching in BOE. A second lift-off and evaporation process (under the same conditions) is then performed to produce the top electrodes including probe contact pads and metal strips connecting relevant vias. Due to the isotropic nature of the BOE process, the vias have slanted sidewalls that ensure the electrical conductance between top and bottom electrodes. The final devices are diced and polished to ensure good coupling from and to optical fibers.
Figure 5.3(a-b) show a range of fabricated nanophotonic LN devices including nano-waveguides, ring resonators, racetrack resonators and MZIs. The typical propagation loss of these structures is \( \sim 3 \text{ dB/cm} \), which is currently limited by etching roughness \((102)\) and can be further improved by at least an order of magnitude \((34, 103)\). The resulting MZI and racetrack structures have low on-chip insertion loss of \( \sim 2 \text{ dB} \) and \( \sim 1 \text{ dB} \) respectively (with additional 5 dB/facet coupling loss).

Figure 5.1 Schematic view of the device layout with thin film LN waveguides and RF electrodes. Metal vias and bridges are fabricated to achieve modulation on both arms of the devices. Inset shows a schematic of the device cross-section with an overview of the metal bridge.
Figure 5.2 Fabrication procedure of the nanophotonic LN modulators

Figure 5.3 Fabricated optical devices and electrical contacts. (a-b) False-color SEM images of the fabricated racetrack and ring resonator based modulators (a) and MZI based modulators (b). (c) Cross-section view of the simulated optical TE mode profile ($E_z$ component) and RF electrical field (shown by arrows). The x-cut LN used here is most sensitive to the horizontal component of the electric field ($E_z$).
The highly confined optical mode allows us to place gold micro-RF electrodes close to the LN waveguide [Figure 5.3(c-d)], enabling low operating voltages. Our devices make use of an x-cut LN configuration, where transverse-electric (TE) optical modes and in-plane electric fields \((E_z)\) interact through the highest electro-optic tensor component \((r_{33})\) of LN. We design the waveguide geometry and the micro-RF electrode positions to achieve the optimal overlap between the optical and electric fields, while minimizing the bending loss and the metal-induced absorption loss. Figure 5.3(c) shows the numerically simulated overlap between the corresponding optical and electric fields. The ridge optical waveguides have a top width \(w = 900\) nm, rib height \(h = 400\) nm, and a slab thickness \(s = 300\) nm [Figure 5.3(c)]. To maximize the in-plane electric field \((E_z)\), we sandwich the optical waveguide between the signal and ground electrodes with a gap \(g = 3.5\) μm. A SiO\(_2\) cladding is used to further enhance this overlap by increasing the dielectric constant of the surrounding media to match the high dielectric constant of LN (\(\epsilon \sim 28\)) (4).
5.2 DC Response of the Nanophotonic Lithium Niobate Modulators

We show efficient and linear electro-optic tuning in a racetrack modulator and a MZI modulator. Light from a tunable telecom laser (Santec TSL-510, 1480 – 1580 nm) is coupled into and collected from the waveguide facets using tapered lensed fibers. A 3-paddle fiber polarization controller is used to ensure TE input polarization. Figure 5.4(a) shows a typical transmission spectrum of a racetrack resonator with a loaded quality (Q) factor ~ 50,000. When a voltage is applied, the change of refractive index modifies the effective optical path length of the resonator, resulting in a resonance frequency shift. The electrical fields on the two racetrack arms are aligned to the same direction so that the modulation on the two arms adds up (Figure 5.1). The measured electro-optic efficiency is 7.0 pm/V with good linearity and no observable changes in resonance extinction ratio and linewidth [Figure 5.4(a-b)]. The MZI modulator is a balanced interferometer with two 50:50 Y-splitters and two optical paths. The applied voltage induces a phase delay on one arm and a phase advance on the other, which in turn change the output intensity at the Y-combiner by interference. The minimum voltage that is needed to completely switch the output between on and off is defined as the half-
wave voltage ($V_\pi$). We measure a $V_\pi$ of 9 V from a 2 mm long MZI modulator, with 10 dB extinction ratio [Figure 5.4(c)]. This translates to a voltage-length product of 1.8 V•cm, an order of magnitude lower than bulk LN devices (85) and significantly better than previously reported LN thin-film devices because of the highly-confined electro-optic overlap (29-31, 34, 38, 40, 100).
Figure 5.4 DC electrical and optical characterization. (a) Measured transmission spectra of a high Q (~ 50,000) racetrack resonator exhibits large frequency shift with applied DC voltage. (b) Linear resonant wavelength shift as a function of DC voltage with error bars. The measured tuning efficiency is 7.0 pm/V. (c) Optical transmission of a 2 mm long MZI modulator versus DC voltage applied, indicating a half-wave voltage ($V_{\pi}$) of 9 V and a voltage-length product of 1.8 V•cm.
5.3 High-Frequency Response of the Modulators

Using the measurement setup shown in Figure 5.5, we characterize the high frequency response of our nanophotonic LN modulators. An erbium-doped fiber amplifier (EDFA) and a band-pass filter (BPF) are used after the chip to increase the optical intensity in the detector. A 50 GHz microwave probe (GGB Model 50A) is used to connect the on-chip metal contacts and the 50 Ω RF coax line. For small-signal bandwidth measurements, a 50 GHz vector network analyzer (VNA, Agilent E8364B) and a 40 GHz photodiode (New Focus 1014) are used. RF cable losses are subtracted from the measured frequency responses. For eye diagram measurements, pseudo-random binary sequence (PRBS, CentellaxTG1P4A) signals are amplified (Centellax OA4MVM3) to 5.66 V peak to peak (V_{pp}) and used to drive the modulators. Eye diagrams are obtained by sending the modulated and amplified light into the 30 GHz optical module of a sampling scope (Tektronix CSA8000). For temperature stability testing, a peltier controller is used to regulate the temperature of the sample stage. During the test, laser wavelength, input polarization and bias voltage are kept constant, while the lensed fiber positions
are fine tuned to compensate for the relative motion between the chip and the fibers.

Figure 5.5 Detailed high-speed measurement setup. VNA and PD (40 GHz) are used for electro-optic bandwidth test. PRBS, RF amplifier and optical sampling scope (with a 30 GHz optical module) are used to obtain the eye diagrams.

Our miniaturized modulators feature high electro-optic bandwidths. For a racetrack resonator modulator featuring a Q factor ~ 8,000, we measure a 3 dB electro-optic bandwidth of 30 GHz [Figure 5.6(a)]. This value is limited by the cavity-photon lifetime of the resonator (~ 6 ps). We confirm the lifetime limited bandwidth by testing additional resonators with Qs of 5,700 and 18,000. The resulting 3 dB bandwidths are 40 GHz and 11 GHz respectively (Figure 5.7). The Q factors are engineered from the intrinsic value by controlling the distance between the RF electrodes and the optical waveguide. The intrinsic RC bandwidth
limit of the racetrack modulator is estimated to be over 100 GHz. For the 2 mm long MZI device with direct capacitive modulation, the measured electro-optic 3 dB bandwidth is ~ 15 GHz [Figure 5.6(b)]. This is limited by the RC constant due to a larger capacitance (~ 0.2 pF) induced by the longer RF electrode used. Note that since our on-chip electrical resistance is rather small (< 10 Ω), the measured bandwidth is currently limited by the 50 Ω impedance of the VNA drive.

Our platform supports data transmission rates as high as 40 Gbps. Figure 5.6(c) displays NRZ open eye diagrams for both racetrack and MZI modulators at various data rates. Because of the high signal quality, our devices can generally operate at data rates 1.5 times their 3 dB bandwidth, which translates to 40 Gbps and 22 Gbps for the racetrack and MZI devices, respectively. At maximum frequencies, the measured extinction ratios of the racetrack and MZI modulators are 3 dB and 8 dB with power consumptions (CV^2/4) of 240 fJ/bit and 1.6 pJ/bit, respectively.

We confirm that our MZI modulator maintains the stable thermal properties of their bulk counterparts, due to the low thermo-optic coefficient of LN (3.9 × 10^{-5} K^{-1}) (4). We vary the temperature of our chip within a ΔT = 20 °C range, limited by the setup, and record the eye diagrams. We find that the MZI modulator is able to
maintain an open eye diagram at the maximum data rate of 22 Gbps without any feedback to compensate for temperature drifts (open loop configuration) [Figure 5.6(e)].
Figure 5.6 Bandwidth and high speed data operation. (a-b) Electro-optic bandwidths ($S_{21}$ parameter) of a race-track resonator with $Q \sim 8,000$ (a) and a 2 mm long MZI (b). (c) Eye diagrams of the racetrack (blue) and MZI (purple) modulator with data rates up to 40 Gbps and 22 Gbps. (d) Eye diagrams of the MZI modulator at 12.5 Gbps and 22 Gbps with the device heated up by 20°C. The extinction ratios in d and e are 3 dB and 8 dB for racetrack resonator and MZI respectively.
Figure 5.7 Measured transmission spectra and electro-optic bandwidths of two representative microresonator modulators, indicating Q factors of ~ 5,700 (a) and 18,000 (c), as well as 3 dB electro-optic bandwidths of ~ 40 GHz (b) and 11 GHz (d).
In recent years, photonic crystal (PhC) cavities have made great progress both theoretically and experimentally (104-108). Thanks to their wavelength-scale modal volumes and high quality factors (Q factors), power and field could be strongly enhanced within these cavities. In particular, PhC nanobeam cavity (PCNC), where light is confined by total internal reflection in two directions, is a promising platform for various applications including optomechanics (109), nonlinear optics (110, 111), biosensing (112) and quantum optics (113). Combining the strong light confinement of PCNCs and the high nonlinearity in LN could enable a range of compact switching and nonlinear optical devices. However, the post-etching non-vertical sidewall discussed in previous chapters prevents the direct implementation of conventional air-hole PCNC designs in our LN platform.
In this chapter, we propose and design a new scheme for LN PCNCs based on periodical nano-grooves that shows promising performance for electro-optic switching and nonlinear wavelength conversions.

The principle of designing an ultra-high Q PCNC is to linearly increase the mirror strength on both sides of the cavity, by carefully engineering system parameters, so as to achieve a Gaussian-like field profile \( (107) \). However, as is mentioned before, the tilted sidewalls caused by dry etching will turn the conventional cylindrical or square air-holes into conical holes, severely reducing the size of the bandgap. To maintain sufficient mirror strength, we use nano-groove structures to replace air-holes, shown schematically in Figure 6.1. The nano-groove structures are constructed such that all sidewalls make an angle \( \theta = 35^\circ \) with respect to the vertical direction, which is determined from our fabrication results. The groove width, depth and period are kept constant over the entire structure, while beam width is quadratically increased. Additional grooves with fixed beam width at both ends of the cavity are used in the TM case.
Figure 6.1 Schematics of photonic crystal nano-groove cavities. (a) Top view. (b) Cross-sectional view on x-z plane.

All simulations in this chapter were performed using 3D FDTD (Lumerical) method. According to the z-cut LNOI wafer we will use in future experiment, here we considered LN as a birefringent crystal with $n_z = n_e = 2.13$ and $n_x = n_y = n_o = 2.21$. And the largest diagonal second order susceptibilities $\chi^{(2)}_{zzz}$ in our case, refers to $r_{33} = 30.9 \text{ pm/V}$ (EO modulation) $d_{33} = 41.7 \text{ pm/V}$ (SHG) (114). Since the thickness of the thin LN film we use (400 nm) is smaller than wavelength, we start with TE modes which possess higher effective indices than TM modes. A typical TE band diagram of a nano-groove PhC is shown in Figure 6.2(a) (red line), with period $a = 450 \text{ nm}$, $b = 50 \text{ nm}$ for a groove width of 400 nm and beam width $W_{\text{min}} = 550 \text{ nm}$. At the edge of the first Brillouin zone, the corresponding frequency is 195 THz (1538 nm) and we take this set of parameters for cavity mode. As the device
extends into the mirror region, the beam width is quadratically increased to \( W_{\text{max}} = 900 \text{ nm} \). In this case no additional grooves are used. The TE band diagram at the end of the mirror region is shown as the black line in Figure 6.2(a), where the operating frequency is right in the center of the bandgap.

Figure 6.2 (a) TE band diagrams for cavity and mirror modes. (b) Theoretical Q factors and modal volumes with different numbers of modulating groove pairs. (c) Ey distribution in x-y plane at the top of the beam. Black lines show the outline of the device.

Figure 6.2(b) demonstrates the Q factors and modal volumes of such cavities with different numbers of mirror groove pairs. As the groove number increases, Q factors grow exponentially and modal volumes grow linearly. With 80 mirror
groove pairs, an ultra-high Q factor of $3.9 \times 10^6$ is achieved. The corresponding modal volume is $5.0 \left(\frac{\lambda}{n}\right)^3$. Figure 6.2(c) shows the corresponding Gaussian-like field distribution of our LN nanobeam cavities. It should be noted that we also simulated the case where the error of tilted angle occurs. Results shows that for the 80 mirror groove pairs cavity, the Q factors could maintain above $3.1 \times 10^5$ and the operating frequency fluctuation is less than 6 THz while the tilted angle error ranges from -5° to 5°.

For nonlinear optical applications using z-cut LNOI substrates, it is appealing to use TM modes, so as to take advantage of the largest nonlinear coefficient component. While TM modes are favored in high-aspect-ratio beams (115), their designs are challenging in a 400 nm thin film since a lower effective index leads to a smaller bandgap. To compensate for the weakened bandgap effect and increase the effective index, the period and the groove width are increased. The final parameters are chosen as: $a = 485$ nm, $b = 50$ nm, $W_{\text{min}} = 470$ nm, $W_{\text{max}} = 900$ nm. The corresponding band diagrams are shown in Figure 6.3(a), indicating an operating frequency of 199 THz (1508 nm).
Figure 6.3 (a) TM band diagrams for cavity, mirror and 2nd order TE modes. (b) Theoretical Q factors, modal volumes and transmissions (T) with different numbers of modulating groove pairs and additional groove pairs (c) $E_z$ distribution in x-y plane at the top of the beam. (d) $E_z$ distribution in x-z plane at the center of the beam. Black lines show the outline of the device.

It should be noted that the 2nd order TE dielectric band [green line in Figure 6.3(a)] is close to the operating frequency and it could cause detrimental loss channels. It shares the same symmetry and has finite field overlap with the 1st order TM mode.
We have carefully avoided the case where the 2nd order TE dielectric band edge intersects with the operating frequency in the mirror region by engineering system parameters. In this case, we achieved a Q factor of $1.8 \times 10^6$ with 100 pairs of modulating grooves. Modal volumes of TM-mode cavities are larger than those of the TE cases, varying from 9 to 15 cubic wavelengths, due to weaker Bragg confinement in $x$ direction and more field leakage in $z$ direction caused by lower effective indices. Figure 6.3(d) shows the field distribution in $x$-$z$ plane. The electric field jumps up as it passes through the top and bottom edges of the beam, following the required boundary conditions. However, most of the energy (> 70%) is still confined in the beam as expected.

Additional fixed-width grooves at each end of the modulating parts could be used to further strengthen the light confinement in $x$ direction and increase the Q factors while maintaining similar levels of modal volumes, shown in Figure 6.3(b). With 70 modulating grooves and 20 additional grooves on each side, the Q factor ($1.5 \times 10^6$) is comparable with the case of 100 modulating grooves, while its modal volume $[11.8 (\lambda/n)^3]$ is 20% smaller. However, the dramatic decrease of waveguide coupling caused by additional grooves will weaken the dominance of waveguide
coupling loss over scattering loss, which is necessary for high transmission (T = 10% for 70 +20 case, as compared with 34% in the 100 + 0 case).

With small footprints and high Q factors, our LN PCNCs could be used to realize compact on-chip EO modulators with low switching voltages. From FDTD simulations, the refractive index sensitivity of our device was calculated to be \( \Delta f/\Delta n = 28.5 \text{ THz/RIU} \) (refractive index unit) or \( \Delta \lambda/\Delta n = 218 \text{ nm/RIU} \). The relation between applied voltage and resulting electric field within the LN beam could be estimated using a parallel capacitor model. By assuming that metal electrodes are placed 0.85 \( \mu \text{m} \) above and 3.65 \( \mu \text{m} \) beneath the LN layer, separated by silicon dioxide layers, which is equivalent to the case reported in the previous LN microring resonator case \( (\varepsilon_{\text{LN}} = 28, \varepsilon_{\text{SiO}_2} = 3.8) \) (29), a frequency tunability of 0.13 GHz/V could be achieved, which is comparable with the theoretical value in (29), but realized within a much smaller footprint. Moreover, the high Q factors significantly reduce the required voltage to switch the cavity between on and off states. In the case of 100 groove pairs \( (Q = 1.4 \times 10^6 \text{ with electrodes}) \), a switching voltage as low as \( \sim 1.4 \text{ V} \) is expected to be achieved. Here in order to eliminate the effects from metal loss, the electrodes are pushed further away (3 \( \mu \text{m} \) silicon dioxide layers on both sides of the device).
The field enhancement in our ultra-high Q nanobeam cavities also leads to a dramatic enhancement in nonlinear optical responses. Herein we take SHG as an example and simulated the SHG response using nonlinear FDTD simulations. A continuous-wave mode source (oscillating at the cavity resonance frequency) was used to pump the system from one waveguide port. Simulation time was set to be sufficiently long so that a steady state was reached with constant output SHG power. The output power was monitored in all directions so as to capture the total internally generated SHG power. Using this method the conversion efficiencies for 40, 45 and 50 mirror groove pairs were calculated and shown as the red stars in Figure 6.4(a).

These results are valid in the limit of low conversion where spontaneous down conversion and pump depletion are negligible. Since the device is singly resonant (SHG wavelength is off resonance), temporal coupled mode theory shows that the conversion efficiency (normalized by input power) is quadratically dependent on the intra-cavity power, or ~ \( (QT^{0.5}/V)^2 \), where T is the cavity transmission and V is the modal volume \((116)\). Using this relation, we numerically predicted the SHG efficiencies for the cases of 45 and 50 mirror pairs [blue spots in Figure 6.4(a)] based on the efficiency of 40 groove pairs, which match well with 3D nonlinear FDTD
simulation results. For groove pair numbers larger than 50, corresponding conversion efficiencies were also predicted and shown in Figure 6.4(a). With a Q factor ~ $10^6$ (100 groove pairs), the total second harmonic power radiated in all directions gives a SHG efficiency of 18.0 W$^{-1}$ theoretically. Figure 6.4(b) shows the farfield analysis of second harmonic radiation from the top of the device. An objective lens with NA = 0.65 would be able to collect 31% of the internally generated SHG power, which corresponds to an efficiency of 5.6 W$^{-1}$.

Figure 6.4 (a) SHG efficiencies for different groove pair numbers using nonlinear FDTD (red stars) and numerical prediction (blue spots). (b) Farfield analysis of scattered SHG light.
Chapter 7

Conclusions and Outlook

For decades, LN has been the material of choice for both nonlinear wavelength conversion and electro-optic modulation. However, due to the difficulty in nanostructuring LN, these devices have remained bulky, power hungry and expensive. We show in this thesis that, by leveraging LN’s superb material properties and the nanofabrication approaches we developed, LN nonlinear photonic devices that are highly efficient, integrable and mass-producible can be realized with standard lithographic processes.

We first demonstrate a nanophotonic LN platform that supports various wavelength-scale devices (e.g. nanowaveguides, microdisk resonators, micro-ring resonators, MZI etc.) with high quality factors (> 100,000). This platform is subsequently used for efficient SHG and high-speed electro-optic modulation. We show that the versatility of our top-down fabrication method provides alternative
phase matching schemes beyond traditional periodic poling approaches, and even the possibility for phase-matching-free nonlinear wavelength generation.

Compared with traditional LN devices that have been developed and optimized for over 40 years, our nanophotonic LN platform has already shown similar or better performance in both wavelength conversion and electro-optic modulation. With more engineering on device design and fabrication, we expect the device performance be further improved by several more orders of magnitude. Below I introduce three major future directions that could potentially reshape the field of nonlinear photonics.

7.1 Fabrication of Ultra-High Quality Photonic Structures

The optical Q factors of our fabricated nanophotonic LN devices are currently limited to ~ 100,000, which translates to a propagation loss ~ 3 dB/cm. Although this number is good enough for proof-of-concept demonstrations, it is lower than typical nanophotonic devices in other material platforms and traditional LN devices (Table 7.1). Further improvement on the Q factors is possible since the material absorption loss of LN is at least two orders of magnitude lower than our current values. Increasing the Q factors by another order of magnitude (i.e. $10^6$)
will enable nonlinear wavelength conversion systems with close-to-unity efficiency and/or single-photon conversion capability, actively tunable on-chip frequency comb sources, as well as electro-optic modulators with ultra-low driving voltages.

Table 7.1 Comparison of optical quality factors/propagation loss achieved in other platforms (~ 1550 nm)

<table>
<thead>
<tr>
<th>Material</th>
<th>Waveguide Type</th>
<th>Q Factor</th>
<th>Propagation Loss</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>Etched</td>
<td>$1.1 \times 10^7$ (117)</td>
<td>3.6 dB/cm (118)</td>
</tr>
<tr>
<td>SiN</td>
<td>Etched</td>
<td>$7 \times 10^6$ (119, 120)</td>
<td>0.003 dB/cm (121)</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>Etched</td>
<td>$8.75 \times 10^8$ (122)</td>
<td>0.045 dB/cm (123)</td>
</tr>
<tr>
<td>AlN</td>
<td>Etched</td>
<td>$6 \times 10^5$ (124)</td>
<td>0.8 dB/cm (124)</td>
</tr>
<tr>
<td>Diamond</td>
<td>Etched</td>
<td>$1 \times 10^6$ (25)</td>
<td>–</td>
</tr>
<tr>
<td>GaN</td>
<td>Etched</td>
<td>$7 \times 10^4$ (125)</td>
<td>–</td>
</tr>
<tr>
<td>SiC</td>
<td>Etched</td>
<td>$1.4 \times 10^5$ (126)</td>
<td>–</td>
</tr>
<tr>
<td>LN (traditional)</td>
<td>Diffused</td>
<td>–</td>
<td>0.3 dB/cm (127)</td>
</tr>
<tr>
<td>LN (bulk)</td>
<td>Polished</td>
<td>$2 \times 10^6$ (128)</td>
<td>–</td>
</tr>
<tr>
<td>LN (this work)</td>
<td>Etched</td>
<td>$1 \times 10^5$</td>
<td>3 dB/cm</td>
</tr>
</tbody>
</table>
Figure 7.1 shows the SEM image of a typical fabricated device with observable sidewall roughness, which is currently the major limitation for propagation loss. The vertical lines on the side walls in Figure 7.1 indicate that the roughness represents a genuine transfer from the a-Si etching mask, which is known to be porous. A different etching mask (e.g., SiO$_2$ or SiN) and further optimization on the resist reflow process could potentially reduce this etching roughness, as people have done in SiN (120).

Figure 7.1 SEM image of a representative device, showing sidewall roughness induced by dry etching process.
7.2 Periodic Poling in Thin-Film Lithium Niobate

In Chapter 3 we introduce two distinct phase matching methods that do not need periodic domain inversion, while keeping a similar level of normalized conversion efficiency as traditional PPLNs. However, these approaches do not take full advantage of the highly-confined LN waveguides. In the case of modal phase matching, the nonlinear overlap between fundamental mode (fundamental harmonic) and 3rd order mode (SH) is much lower than that between fundamental modes at both wavelengths. In the case of PGLN, the trade-off between nonlinear overlap and propagation loss always exists.

Going forward, periodic poling in thin-film LN could be used to leverage both LN’s excellent material property and the strong light-matter interaction enabled by our nanowaveguides (Figure 7.2). Recently, SHG has been demonstrated in thin-film PPLN, where waveguiding is achieved by loading an index matching rib (SiN) on top of the thin film (39). Our simulation shows that, periodic poling in dry etched LN nanowaveguides could further improve the nonlinear overlap, with theoretical SHG conversion efficiency > 6,000% W⁻¹cm² (Figure 7.3). This value represents two orders of magnitude improvement over traditional PPLN
and could enable efficient quantum wavelength conversion. The challenge in realizing thin-film PPLN lies in the short poling periods required by the high-confinement waveguides. The strong waveguide dispersion in sub-wavelength waveguides results in QPM periods \( \sim 3 \, \mu\text{m} \) (Figure 7.3), much shorter than traditional poling periods \((10 \sim 20 \, \mu\text{m})\) \((61, 127, 129-137)\).

Figure 7.2 Schematic of the proposed thin-film PPLN platform. Sub-micron x-cut LN thin film is first periodically poled in-plane in regions of interest, and then dry-etched to realize waveguides and resonators with engineered dispersion and phase matching.
Figure 7.3 Calculated QPM periods (solid lines) and normalized conversion efficiencies (dotted lines) for thin-film PPLN (400 nm thick) waveguide with rectangular (blue) and trapezoidal (red) cross-section. The latter, motivated by the profile of fabricated devices, features dip at ~1.4 μm width which is due to avoided mode crossing between 1\textsuperscript{st} TE and 2\textsuperscript{nd} TM modes.

7.3 CMOS-Drivable Electro-Optic Modulators

The switching voltages of traditional LN modulators are 5 ~ 7 V, usually supplied by expensive and power-consuming electrical amplifiers. The proof-of-concept nanophotonic modulator (MZI type) discussed in Chapter 5 features an electro-optic figure of merit of 1.8 V•cm, which means that a nanophotonic LN modulator with 2 cm device length can be switched between on and off states by a sub-volt
external field. Such modulators could potentially be directly driven by CMOS circuitry and are particularly promising for data center applications whose power consumption is posing increasing challenge.

In order to achieve this performance benchmark, a two-fold optimization to our current modulators is required. First, the waveguide propagation loss over the 2 cm device needs to be negligible (i.e. < 1 dB/cm), as is discussed in Section 7.1. Second, an RF transmission line that is phase-matched with the optical light wave needs to be implemented, instead of the direct capacitive modulation in our current devices that limits the electro-optic bandwidth.

Such RF transmission line is possible and has already been demonstrated in thin-film LN platforms with an index-matching rib waveguide architecture (40, 100). In fact, the existence of buried and cladding oxide offers unique advantage in designing a transmission line that is both phase matched with optics and impedance matched with the RF network. For over a decade, existing LN modulator performance has been capped due to the nonideal phase matching between the RF and optical fields (85). The high dielectric constant of LN ($\varepsilon_{RF} \sim 28$) (4) dictates that RF fields in LN propagate much slower than optical fields ($\varepsilon_{opt} \sim 4$), resulting in performance trade-off between bandwidth and driving voltage. In
our thin-film LN approach, instead, phase matching can be much better achieved since the electrical field primarily resides in the low dielectric SiO2 substrate ($\varepsilon_{\text{RF}} \sim 4$) and readily propagates at nearly the same group velocity as light ($85$). With a phase-matched RF transmission line, our thin-film micro-MZI modulator could simultaneously achieve ultra-high bandwidth (> 60 GHz) and low modulation voltage (< 1 V).

Table 7.2 shows a comparison of modulator performance between other competing platforms, our current results, and the proposed scheme with RF transmission line. This high-performance monolithic LN nanophotonic platform could become a practical cost-effective solution to meet the growing demands of next-generation data centers and long-haul optical telecommunications.
Table 7.2 Performance comparison between our nanophotonic LN modulators and other competing platforms

<table>
<thead>
<tr>
<th>Figures</th>
<th>Bulk LN</th>
<th>Si</th>
<th>InP</th>
<th>This work</th>
<th>Proposed traveling wave</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bandwidth</td>
<td>40 GHz</td>
<td>30 GHz</td>
<td>30 GHz</td>
<td>30 GHz</td>
<td>60 GHz</td>
</tr>
<tr>
<td>Voltage ($V_{π}$)</td>
<td>7.5 V</td>
<td>6 V</td>
<td>1.5 V</td>
<td>~5 V</td>
<td>1 V</td>
</tr>
<tr>
<td>Insertion loss</td>
<td>3 dB</td>
<td>15 dB</td>
<td>10 dB</td>
<td>12 dB</td>
<td>&lt; 3 dB</td>
</tr>
<tr>
<td>Linearity</td>
<td>Linear</td>
<td>Nonlinear</td>
<td>Nonlinear</td>
<td>Linear</td>
<td>Linear</td>
</tr>
<tr>
<td>Size</td>
<td>&gt; 10 cm</td>
<td>20 µm</td>
<td>1 cm</td>
<td>40 µm</td>
<td>1 cm</td>
</tr>
</tbody>
</table>


91. M. Aoki et al., InGaAs/InGaAsP MQW electroabsorption modulator integrated with a DFB laser fabricated by band-gap energy control selective area MOCVD. IEEE J. Quant. Electron. 29, 2088-2096 (1993).


