Integrated Nanophotonic Devices in Lithium Niobate

Mohammad Amin Baghban
Front Cover:
*A photonic integrated circuit containing superconducting single-photon detectors on thin-film lithium niobate optical circuitry.*

KTH Royal Institute of Technology
School of Engineering Sciences
Department of Applied Physics
Quantum and Biophotonics unit

TRITA-SCI-FOU 2018:44

Akademisk avhandling som med tillstånd av KTH i Stockholm framläggdes till offentlig granskning för avläggande av teknisk doktorsexamen fredagen den 26:e oktober klockan 10:00 i sal FA32, KTH, Roslagstullsbacken 21, Stockholm.

© Mohammad Amin Baghban, October 2018.

Tryck: Universitetetsservice US AB
Dedicated to

my parents

—for their never-ending love—

and to people striving for justice and freedom
Abstract

Lithium niobate (LN) is a ferroelectric crystal offering a broad transparency spectrum, together with excellent electro-optic and nonlinear optical properties. Thanks to them, LN is setting the standard for quantum optics and telecommunications in critical applications such as ultrafast modulation and frequency conversion. The development of a reliable nanophotonic platform in LN can be expected to effectively leverage all such appealing functionalities in compact and integrated formats and provide important and complementary functionalities to current silicon-photonics platforms.

This thesis encompasses systematic and consistent efforts with the goal to achieve the key building blocks for a comprehensive integrated nanophotonic platform in LN. It involves work on the technology side, sustained and complemented by modelling and experiments, ultimately leading to the demonstration of a few novel devices.

Ultrahigh field confinement in nanophotonic waveguides is accompanied by the appearance of non-negligible longitudinal components in the guided optical fields. By fabricating high-quality LN nanopillars and analyzing with theory and experiments their second harmonic generation (SHG) response, we provide evidence for the existence of longitudinal field components and demonstrate the possibility to reshape the SHG polar emission properties of these submicrometric waveguides by fine-tuning the nanopillar size.

This thesis also presents a different technological approach, allowing the fabrication of photonic wires as small as 250 nm with lengths up to 1 cm on LN-on-insulator (LNOI), suitable for upscaling to photonic integrated circuit (PIC) architectures. By optimizing the fabrication process, the propagation losses of single-mode waveguides at telecom wavelengths on this platform were brought down from 76 to 1.13 dB/cm. Fine-pitch waveguide structuring was also successfully achieved, enabling LNOI-to-fiber grating couplers and waveguide Bragg gratings, the latter featuring record extinction ratios in LNOI (45 dB), comparable to the state of the art in silicon.

The thesis involves also theoretical work on the design of photonic wires where the interplay between LN and waveguide birefringence is used to achieve polarization-insensitive operation for the fundamental guided modes.

Finally, two demonstrators are provided for novel and emerging applications of LN to the life sciences, using LNOI surface-patterned templates for enhanced Raman spectroscopy and LN templates for controlled neuron growth and manipulation in microfluidic environments, respectively.

**Keywords:** Lithium niobate (LN), LN-on-insulator, photonic integrated circuits, nonlinear optics, electro-optics, waveguides, propagation losses, Bragg gratings, grating couplers, polarization-insensitive, LN for life sciences.
Sammanfattning

Litium niobat (LN) är en ferroelektrisk kristall som erbjuder ett brett transparent spektrum tillsammans med utmärkta elektrooptiska och ickelinjära optiska egenskaper. Tack vare dem sätter LN standarden för kvantoptik och telekommunikation i kritiska applikationer som ultrasnabb modulering och frekvensomvandling. Utvecklingen av en pålitlig nanofotonikplattform i LN kan förväntas effektivt utnyttja alla sådana tilltalande funktioner i kompaktera och integrerade format och ge viktiga och komplementära funktioner till nuvarande plattformar för kiselfotonik.

Denna avhandling omfattar systematiska och konsekventa insatser med målet att uppnå de viktigaste byggnestenarna för en omfattande integrerad nanofotonikplattform i LN. Det handlar om arbete på teknisksidan, som upprätthålls och kompletteras med modellering och experiment, vilket leder till demonstrationer av några nya enheter.

Ultrahög fältpåverkan i nanofotoniska vågledare åtföljs av uppkomsten av icke försämrbara längsgående komponenter i de guidade optiska fälten. Genom att tillverka högkvalitativa LN nanopelare och analysera med teorin och experimentellt undersöka deras andra harmoniska generationssvar (SHG), ger vi bevis för förekomsten av longitudinella fältkomponenter och visar möjligheten att omforma SHG-polära utsläppsegenskaperna hos dessa submikrometriska vågledare genom finjustering av deras storlek.

Denna avhandling presenterar också ett annat tekniskt tillvägagångssätt som gör det möjligt att tillverka fotoniska vågledare så små som 250 nm med längder upp till 1 cm på LN-på-isolatorn (LNOI), som är lämpliga för uppskalning till fotoniska integrerade kretsarkitekter (PIC). Genom optimering av tillverkningsprocessen sänktes förökningsförlusterna av singelmodsvågor vid telekomvåglängder på denna plattform ned från 76 till 1,13 dB/cm. Strukturering av LNOI-vågledare med fina tonhöjder uppnådes också framgångsrikt, vilket möjliggjorde LNOI-till-fiber-gitterkopplingar och vågledar Bragggitter, den senare med rekordutrotningsförhållanden i LNOI (45 dB), jämförbar med toppmodern i kiselfotonik.

Avhandlingen innefattar också teoretiskt arbete med utformning av fotoniska vågledare där samspelet mellan LN och vågledarförsjutning används för att uppnå polarisationsinsensitiv drift för de grundläggande styrda lägena.

Slutligen tillhandahålls två demonstranter för nya och framväxande applikationer av LN till biovetenskap, med hjälp av LNOI ytmönstrade mallar för förbättrade Raman spektroskopi och LN mallar för konsenerad neurontrav och manipulation i mikrofluidiska miljöer.
Acknowledgements

I would therefore like to take the chance here to acknowledge help and support from the many individuals whose assistance has eased the accomplishment of this thesis.

My deep, sincere and thankful gratitude goes to Prof. Katia Gallo, my main supervisor, for her supervision and help while letting me steer my research, understanding and friendship. Katia! I definitely cannot value what I have learned from you (scientifically and personally) since I entered KTH. It has been an immense privilege to work with you!

Many thanks to my co-supervisor, Prof. Gunnar Björk. Thanks for accepting me to QEO. Your unbounded experiences have been very instructive for me and I admire your deep and broad scientific knowledge.

Thanks to Dr. Marcin Swillo for his kind advises, personally and scientifically, and for all I learned from him. Thanks for your friendship and for letting me use your setup for the experiments.

And my sincere and respectful gratitude to Prof. Brian J. Rodriguez! I learned a lot through our fruitful collaboration and your scientific character. I hope we can keep working together!

I would also like to thank Dr. Max Yan for being my advanced thesis reviewer. My special thanks go to Dr. Adrian Iovan, Dr. Anders Liljeborg and Dr. Anders Holmberg for kindly helping me to use the cleanroom, and for managing Albanova NanoFabLab cleanroom, specifically Dr. Adrian Iovan for his friendship. I also sincerely thank Dr. Lech Wosinski for letting me (and other students within KTH) freely use his lab.

My current knowledge in optical sciences is significantly beholden to my previous experiences. My deep appreciations and admirations belong to Dr. Alireza Gharavi, my MSc supervisor in Shiraz University, Iran. Thanks for letting me work within your group, for allowing us to –literally– live in Photonics Lab, for being there all the time for us, and for your kindness. I also express my sincere thanks to Prof. Sasan Fathpour, my former supervisor at CREOL, USA. I undeniably learned much working with you, both scientifically and personally.

To my close friends, Reza Zandi-Shafagh and Yashar Hormozan! I cannot thank you enough for your friendship and company, and for letting me jump into your mini-gang! Thanks for all the time we spent together, for being there for me and for helping me with whatever I had, and I was. Your honest and supportive friendship is among my biggest achievements in Sweden. I further express my gratitude to our mutual friends, Mahdi Moeen, Arsalan Jami and Dr. Amir Djalalian for their friendship and company.

Thanks to my friends in the former QEO (now QEO+NQP) for their sincere friendship and making such a beautiful atmosphere in Albanova. Special
thanks to Dr. Jonas Almlöf for being my friend, and for keeping in touch with me. It truly means a lot to me! Thanks to my dear friend, Eleonora De Luca for her honest friendship, all coffee breaks we had together, and whatever daily-life we talked about. I express my gratitude to Dr. Maryam Sanaee, my old friend and new office-mate, for her sincere friendship, kindness and support. And of course, I need to thank my good friend, Jean Schollhammer for coping with me as his office-mate, for countless lunches and drinks we had together, and for the random topics we talked about. I acknowledge Dr. Michele Manzo for helping me to get to know KTH and Sweden routines when I just came to KTH, and for all random discussions we had when we shared the office. Thanks to Nicklas Bjärnhall Prytz for his friendship and renewal of atmosphere since he joined NQP. And many thanks to my former and current colleagues here: Katarina Stensson, Ömer Bayraktar, Anton Möller, Dr. Saroosh Shabbir, Dr. Aziza Sudirman and Mattias Jönsson for making Albanova such a peaceful environment.

I would also like to thank the many friends I have in QNP. Thanks to Thomas Lettner for being such a supportive, kind and caring friend. I thank my dear friends, Katharina Zeuner and Lucas Schweickert, for their never-ending kindness. Thanks to Julien Zichi, Samuel Gyger and Dr. Ali Elshaari, for their friendship and working with me. I thank Dr. Klaus Jöns for his friendship and all I learned from him, and my friends, Dr. Lily Yang and Dr. Stephan Steinhauer, for helping me with any questions I had. I also thank my friend, Rene Van Der Molen, for all outside-university time we spent together.

Furthermore, I would like to thank my friends within KTH. Thanks to Carlos Errando-Herranz for his friendship, ideas and collaborating with me. Thanks to Dr. Xu Sun, Erik Holmgren, Dr. Joonas Govenius, Hoda Kianirad, Dr. Karolis Parfeniukas and Dr. Riaan Coetzee for their friendship and working together with me.

The last but the most important, I would like to use the chance here to thank my lovely family for their endless support through my whole life, in whatever stage I was and whatever I did. Thanks forever to my parents for their unique kindness and care all the time. Sincere gratitude to my peerless brother, Fahim, for his never-ending sympathy, support and care. And to azize dele man, my sister Hanieh, for her kind-heartedness and care all the time!
# Contents

Abstract ........................................................................................................................................................................... v  
Sammanfattning ................................................................................................................................................................ vi  
Acknowledgements ............................................................................................................................................................. vii  
Contents .............................................................................................................................................................................. ix  
List of Figures .................................................................................................................................................................... xi  
List of Tables ..................................................................................................................................................................... xvi  
List of publications ............................................................................................................................................................. xvii  
Contributions to the papers ................................................................................................................................................ xviii  
Publications not included in this thesis .................................................................................................................................... xix  
Conference Contributions ................................................................................................................................................... xx  
List of Acronyms ............................................................................................................................................................... xxi  
List of symbols .................................................................................................................................................................. xxii  
1 Introduction ............................................................................................................................................................................ 1  
1.1 Background .................................................................................................................................................................... 1  
1.2 Objectives and main achievements ...................................................................................................................................... 4  
1.3 Structure of the thesis ..................................................................................................................................................... 6  
2 Properties of lithium niobate ................................................................................................................................................. 9  
2.1 Crystalline structure ............................................................................................................................................................. 9  
2.2 Dielectric permittivity .......................................................................................................................................................... 10  
2.2.1 Refractive index ........................................................................................................................................................... 11  
2.3 Electro-optics ....................................................................................................................................................................... 13  
2.4 Nonlinear optics ................................................................................................................................................................... 14  
2.5 Ferroelectricity .................................................................................................................................................................... 15  
2.5.1 LN templates for cell growth patterning .......................................................................................................................... 16  
2.5.2 LN templates for photodeposition ................................................................................................................................... 16  
2.5.3 LN templates for Raman enhancement ............................................................................................................................ 16  
3 Theory of guided wave devices .............................................................................................................................................. 19  
3.1 Optical waveguides ............................................................................................................................................................ 19  
3.1.1 Maxwell’s equations and guided modes .......................................................................................................................... 20  
3.1.2 Waveguide dispersion .................................................................................................................................................... 23  
3.1.3 Waveguide birefringence ............................................................................................................................................... 24  
3.1.4 Coupled-mode equations in waveguide configurations ................................................................................................... 24  
3.2 Waveguide gratings ............................................................................................................................................................. 25
List of Figures

Figure 2.1. (a) Schematic representation of the LN crystal structure; (b) the displacement of the Li⁺ and Nb⁵⁺ ions with respect to the O²⁻ planes induces a spontaneous polarization, $P_s$, along the Z axis; (c) schematic representation of the three-fold symmetry of LN: the mirror-symmetry planes ($Y$-$Z$ axes) are indicated by solid blue planes. Subset indicates a possible combination of ($X$, $Y$, $Z$) axes with respect to symmetry planes (solid blue lines). .......... 10

Figure 2.2. Ordinary (solid blue curve) and extraordinary (dashed red curve) refractive indices of LN at room temperature, as a function of wavelength. .................. 11

Figure 2.3. Schematic representation of domains with opposite polarities in a PPLN structure. Positive (green) and negative (red) bound charges at ferroelectric interfaces (dashed circles) are also depicted. .................................................. 15

Figure 3.1. Some commonly used geometries to build optical waveguides: (a) circular, (b) slab, and (c) ridge waveguides. $z$ is the propagation direction .................. 20

Figure 3.2. Waveguide dispersion in a 1 µm-wide and 300 nm-thick LNOI waveguide, for the two lowest-order guided modes: effective indices of the $TE_{00}$ (solid blue line) and $TM_{00}$ (solid red line) modes plotted as a function of wavelength. The dashed black line represents the refractive index of SiO₂, i.e. its intersection with the solid curves corresponds to the cut-off condition of the two guided modes (see eq. 3.14). Simulations performed with COMSOL Multiphysics®. .................................................. 24

Figure 3.3. (a) Codirectional, (b) contradirectional coupling of guided modes by means of a grating. (c) Coupling of a radiation mode to a guided mode $\mu$, where $\beta_\mu$ is the $z$ component of propagation vector of free-space mode ........... 26

Figure 3.4. (a) An example of how two electrodes with a spacing of $G$ can be used to apply an electric field to a $Y$-cut, $X$-propagating LNOI waveguide. (b) Simulated transverse ($Y$, $Z$) distribution of the normalized electric field of the $TE_{00}$ guided mode, $E_{opt}$, at optical frequencies, in the waveguide cross section. The 300 nm-thick waveguide has a trapezoidal cross-section, with a width of 800 nm on top. (c) Simulated distribution of the normalized electric field amplitude (color scale) and field lines (white vectors), produced by an external voltage $V$ applied to the lateral electrodes. $G = 5 \mu$m and scale bars denote a length of 1 µm. Simulations performed using a COMSOL Multiphysics® FEM software .......................................................... 28

Figure 3.5. Collinear interaction of a generic guided optical mode of order $\mu$ with frequency $\omega_{off}$ with a second harmonic guided mode of order $\nu$ and frequency $\omega_{shi} = 2\omega_{off}$, supported by a waveguide with a second-order optical nonlinearity ($\chi^{(2)}$). .............................................................. 29

Figure 4.1. Flow-charts for the different fabrication processes employed in this thesis. (a) Nanopillar fabrication by focused ion beam milling; (b) waveguide
fabrication on LNOI by electron-beam lithography and etching; (c) periodic poling of bulk LN. Pictures: (a) scanning electron microscopy image of a fabricated nanopillar, (b) atomic force microscopy image of a fabricated waveguide grating, (c) piezo-response force microscopy image of two-dimensional hexagonal ferroelectric domains in a Z-cut PPLN crystal.  

**Figure 4.2.** Examples of how different parameters of the ion beam can affect the quality of LN nanopillars fabricated by FIB milling: (a) serpentine vs. (b) raster scan, and raster scan with (c) 1 µs and (d) 5 µs dwell times. Nanopillars made with an ion beam current of 30 pA. Scale bars represent a length of 1 µm.  

**Figure 4.3.** Fabrication steps in the realization of LNOI integrated optical devices: (a) diced thin-film LNOI-on-silicon chip, (b) Chromium hard mask deposition, (c) spin-coating and soft-baking of a resist layer, (d) lithography and pattern-transfer by dry etching of Cr, (e) dry etching of LN, (f) removal of the Cr mask. The arrows indicate the crystalline axes of the LN thin film. \( w_0 \) is the width of the fabricated LN feature (e.g. waveguide).  

**Figure 4.4.** Schematic representation of an EBL system. Inset: illustration of the results of exposure with a given e-beam pattern on a positive (top) and negative (down) resist (in grey).  

**Figure 4.5.** (a) Schematic representation of an ICP-RIE architecture, and schematic illustrations of (b) isotropic and (c) anisotropic etching profiles through the opening in an etch mask (in grey). The arrows indicate etching direction.  

**Figure 4.6.** Some dry etching results: (a) etched hard mask in chromium. Etching results for LNOI waveguides made by photolithographic patterning: (b) LN dry-etched in SF₆/Ar, with evidence of LiF redeposition on the waveguide walls ((image taken before Cr removal), (c) LN dry-etched as in (b), but with an additional wet-etch step to remove the LiF layer. Results for EBL-patterned structures in LNOI: (d) waveguide etched in a SF₆/Ar gas mixture and (e) waveguide and sidewall grating etched by Ar-ion milling, and (f) smooth waveguide sidewalls obtained with the final an optimized Ar-ion milling recipe. Scale bars represent lengths of 1 µm.  

**Figure 4.7.** Metal patterning by a lift-off process. (a) Resist patterning with undercut; (b) uniform deposition of a metal layer thinner than the resist and (c) selective liftoff of the metal layer upon removal of the underlying resist. (d) SEM image of waveguide Bragg grating with gold side-electrodes patterned by EBL and lift-off.  

**Figure 4.8.** (a) Schematic diagram of the poling electrical setup. (b) Sketch of poling configuration with electrical contacts on LN made through the openings of a patterned electrically-insulating resist layer. (c) Typical voltage and current waveforms for the periodic poling of a 0.5mm-thick congruent LN crystal in the self-termination regime.
Figure 4.9. Optical setup used for butt-coupling 1550 nm-light into LNOI nanowaveguides, with lensed fibers and microscope objectives. ................................................. 47

Figure 4.10. (a) Optical setup used for transmission measurements using grating couplers. (b) Sketch of the planar waveguide geometries on chip, including in/out couplers, tapers and waveguide Bragg gratings. (c) Sketch of the configuration used for out-of-plane coupling from a standard telecom optical fiber into the LNOI waveguide via a grating coupler. (d) SEM image of a LNOI grating coupler ................................................................. 48

Figure 4.11. (a) Fabry-Perot measurement (waveguide throughput as a function of wavelength) for a 9 mm-long, 2.9 µm-wide and 300 nm-thick LNOI waveguide. (b) Logarithmic plots of the transmission measured for different waveguide lengths (markers) and linear fits (dashed lines) used to extrapolate their propagation losses by the cut-back method. Data for 300 nm-thick LNOI waveguides with widths of 0.66 µm and 2.35 µm are plotted in orange and green, respectively. ................................................................. 49

Figure 4.12. (a) Sketch of the experimental setup used for polarimetric SHG measurements, with indication of the NP orientation with respect to the LN crystal-axes of LN (X, Y, Z). Example of the polarimetric response retrieved by: (b) simulations of nonlinear interactions in the nanopillars (own MATLAB code) and (c) experiments, here shown for the case of X-polarized SH in bulk Z-cut LN, with a varying in-plane (X-Y) polarization of the FF pump, θ being the FF polarization angle with respect to the X axis of the crystal ........................................................................................................ 50

Figure 5.1. Tilted (52◦) SEM image of a 325 nm NP in Z-cut LiNbO3. Examples of calculated transverse (x, y) distributions of the transverse (E_t) and longitudinal (E_z) field components supported by a 275 nm NP; (b) HE_{11}^{(X)} mode at λ_{FF} = 850 nm; (c) HE_{11}^{(X)}, TE_{01}, HE_{21}^{(X)}, and TM_{01} modes at λ_{SH} = 425 nm. (d) Theoretical polar plots of X-polarized SH intensity in a Z-cut NP as a function of the input FF polarization angle θ (see the inset), calculated for the two limit cases of purely transverse (red) and longitudinal-transverse (blue) SHG coupling. Experimental Polar plots for X-polarized SHG in Z-cut NPs with diameters of (c) 275 nm and (g) 175 nm show how polarimetric response of each NP can change. Solid lines: numerical fits based on Eq. (2) in Paper A. ........................................................................................................ 55

Figure 5.2. Schematic illustration of interplay between waveguide birefringence (left) and intrinsic birefringence of LN (right) can combine to obtain birefringence-free waveguides proposed in Paper B ........................................................................................................ 56

Figure 5.3. Regions in the two-dimensional parameter space defined by wavelength and waveguide width, where the difference of the TE_{00} and TM_{00} mode effective (blue) and group (red) indices is below 0.001 for: (a) ridge and (b) buried waveguides in 300 nm-thick Y-cut LNOI. Plots reprinted from [133]. ............. 57
Figure 5.4. SEM images of (a) a sinusoidal sidewall grating of period XXX, made in a waveguide of average width \( w_0 = 760 \text{ nm} \), with a width modulation depth \( \Delta w = 125 \text{ nm} \), and (b) a rectangular sidewall grating of the same period, made in a waveguide of average width \( w_0 = 700 \text{ nm} \) width a width modulation depth \( \Delta w = 125 \text{ nm} \). The scale bars represent a length of 1 \( \mu \text{m} \).

Figure 5.5. Normalized transmission spectra of rectangular Bragg gratings measured of different values of the width corrugation, \( \Delta w \) (50, 75, 100 and 125 nm, color-coded in the legend). (a) detection-limited data published in Paper C (experiments in 2017) and (b) experimental results obtained from the same sample after improving the measurement sensitivity, showing contrast ratios up to xx in the Bragg grating response.

Figure 5.6. (a) Fiber-to-fiber spectral transmission response of a 0.86 \( \mu \text{m} \)-wide, 500 nm-thick, 0.5 mm-long waveguide; (b) logarithmic plots of the transmission measured for waveguides similar to that of part (a), but with different lengths (markers) and linear fits (dashed line) used to extrapolate their propagation losses by the cut-back method. Values for markers and error bars are obtained by calculating average and standard deviations of transmission response of each waveguide at the spectral range of 1550±20 nm.

Figure 5.7. (a) PIERS in LNOI platform: pre-irradiation of the LN patterned sample creates photogenerated carriers at the LN surface which can be transferred to Ag nanoparticles and enhance the Raman signals. (b) Roughness of the etched LN surface may further enhance the scattered signal. (c) Dielectric interfaces in an LNOI sample can be employed to enhance Raman response at specific wavelengths.

Figure 5.8. (a) Raman spectra of an organic molecule on X-cut LNOI (green) and X-cut LN (black) showing the enhancement associated to the optical multilayered structure of the LNOI template; (b) background-subtracted intensity for the 1583 cm\(^{-1}\) band for in-situ radiation for the organic molecule on Ag-deposited etched X-cut LNOI (black) and Ag-deposited unetched Y-cut LNOI (red). Reprinted with permission from R. Al-Shammari \etal, “Photo-induced enhanced Raman from lithium niobate on insulator template”, ACS Applied Materials & Interfaces 10(36), 30871-30878 (2018). Copyright 2018 American Chemical Society.

Figure 5.9. Alignment of neuronal axons (fluorescently labelled in the confocal images) with topographic features of selectively etched LN substrates in: (a) 9 mm-long micro-grooves (horizontal lines in the picture), and (b)-(c) hexagonal pits (whose edges are marked in green). Images reprinted from Paper E. Reprinted with permission from R. D. Kilinc \etal, “Charge and topography patterned lithium niobate provides physical cues to fluidically isolated cortical axons”, Applied Physics Letters 110(5), 053702 (2017). Copyright 2017 American Institute of Physics.
Figure 6.1. SEM images of 2D arrays of 1 µm-long nanopillars in LN, with a period of 850 nm and NP diameters of (a) 350 nm and (b) 250 nm, fabricated by FIB milling. 1 µm scale bar........................................69

Figure 6.2. SEM images of (a) ring, and (b) racetrack resonators fabricated by the refined EBL and Ar-beam milling process of this thesis in LNOI. The scale bars correspond to 20 µm...............................70
List of Tables

Table 5.1. Examples of extinction ratios achieved in waveguide Bragg gratings fabricated over LN platform. .................................................................58
List of publications

This thesis is based on the following peer-reviewed journal articles:

**Paper A**
M. A. Baghban, K. Gallo, Impact of longitudinal fields on second harmonic generation in lithium niobate nanopillars, APL Photonics 1(6), 061302 (2016).

**Paper B**

**Paper C**

**Paper D**

**Paper E**
Contributions to the papers

**Paper A**
I fabricated the nanopillars, performed experiments and simulations, developed the theoretical model and wrote the paper under the supervision of K. Gallo.

**Paper B**
I supervised J. Schollhammer during his MSc project. I actively contributed to the discussions on the project and paper, and also in writing the paper.

**Paper C**
I fabricated the samples, performed all the experiments, the data analysis and simulations on the waveguide Bragg Gratings and led the writing of the paper.

**Paper D**
I designed and fabricated the samples, participated to the scientific discussions and analysis of the experimental results and actively contributed to writing the manuscript.

**Paper E**
I designed and manufactured the samples, ad-hoc for the needs of our collaborators in Ireland, discussed the results, and actively contributed to writing the manuscript.
Publications not included in this thesis

**Paper F**

**Paper G**
Conference Contributions

Peer-reviewed conferences


Regional conferences


## List of Acronyms

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>AFM</td>
<td>Atomic Force Microscopy</td>
</tr>
<tr>
<td>EBL</td>
<td>Electron Beam Lithography</td>
</tr>
<tr>
<td>EO</td>
<td>Electro-Optic</td>
</tr>
<tr>
<td>FBMS</td>
<td>Fixed-Beam, Moving-Stage</td>
</tr>
<tr>
<td>FEM</td>
<td>Finite-Element Method</td>
</tr>
<tr>
<td>FIB</td>
<td>Focused Ion Beam</td>
</tr>
<tr>
<td>FOM</td>
<td>Figure of Merit</td>
</tr>
<tr>
<td>FWHM</td>
<td>Full-Width-at-Half-Maximum</td>
</tr>
<tr>
<td>ICP-RIE</td>
<td>Inductively Coupled Plasma-Reactive Ion Etching</td>
</tr>
<tr>
<td>LN</td>
<td>Lithium Niobate</td>
</tr>
<tr>
<td>LNOI</td>
<td>Lithium Niobate on Insulator</td>
</tr>
<tr>
<td>NLO</td>
<td>Nonlinear Optic</td>
</tr>
<tr>
<td>PFM</td>
<td>Piezo-Response Force Microscopy</td>
</tr>
<tr>
<td>PIC</td>
<td>Photonic Integrated Circuit</td>
</tr>
<tr>
<td>PIERS</td>
<td>Photo-Induced Enhanced Raman Spectroscopy</td>
</tr>
<tr>
<td>PPLN</td>
<td>Periodically-Poled Lithium Niobate</td>
</tr>
<tr>
<td>RIE</td>
<td>Reactive Ion Etching</td>
</tr>
<tr>
<td>SEM</td>
<td>Scanning Electron Microscopy</td>
</tr>
<tr>
<td>SERS</td>
<td>Surface-Enhanced Raman Spectroscopy</td>
</tr>
<tr>
<td>SH</td>
<td>Second-Harmonic</td>
</tr>
<tr>
<td>SHG</td>
<td>Second Harmonic Generation</td>
</tr>
<tr>
<td>SiN</td>
<td>Silicon Nitride</td>
</tr>
<tr>
<td>SOI</td>
<td>Silicon-on-Insulator</td>
</tr>
<tr>
<td>SPM</td>
<td>Scanning Probe Microscopy</td>
</tr>
<tr>
<td>SSPD</td>
<td>Superconducting Single Photon Detectors</td>
</tr>
<tr>
<td>TE</td>
<td>Transverse Electric</td>
</tr>
<tr>
<td>TEM</td>
<td>Transverse Electro-Magnetic</td>
</tr>
<tr>
<td>TM</td>
<td>Transverse Magnetic</td>
</tr>
<tr>
<td>TMM</td>
<td>Transfer Matrix Method</td>
</tr>
<tr>
<td>UV</td>
<td>Ultra-Violet</td>
</tr>
</tbody>
</table>
## List of symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>Magnetic flux density</td>
</tr>
<tr>
<td>c</td>
<td>Light propagation speed in vacuum</td>
</tr>
<tr>
<td>D</td>
<td>Electric flux density</td>
</tr>
<tr>
<td>d</td>
<td>Second-order nonlinearity coefficient</td>
</tr>
<tr>
<td>E</td>
<td>Electric field</td>
</tr>
<tr>
<td>$E_{opt}$</td>
<td>Transverse distribution of the electric field of an optical mode</td>
</tr>
<tr>
<td>H</td>
<td>Magnetic field intensity</td>
</tr>
<tr>
<td>K</td>
<td>Grating vector</td>
</tr>
<tr>
<td>n</td>
<td>Refractive index</td>
</tr>
<tr>
<td>$n_e$</td>
<td>Extraordinary refractive index</td>
</tr>
<tr>
<td>$N_{eff}$</td>
<td>Effective refractive index</td>
</tr>
<tr>
<td>$N_g$</td>
<td>Group index</td>
</tr>
<tr>
<td>$n_o$</td>
<td>Ordinary refractive index</td>
</tr>
<tr>
<td>P</td>
<td>Polarization</td>
</tr>
<tr>
<td>P</td>
<td>Power</td>
</tr>
<tr>
<td>r</td>
<td>Electro-optic coefficient</td>
</tr>
<tr>
<td>R</td>
<td>Reflection coefficient</td>
</tr>
<tr>
<td>$S_{eff}$</td>
<td>Effective cross section of the nonlinear interaction</td>
</tr>
<tr>
<td>$u_e$</td>
<td>Energy density of the electric field</td>
</tr>
<tr>
<td>$v_g$</td>
<td>Group velocity</td>
</tr>
<tr>
<td>$v_p$</td>
<td>Phase velocity</td>
</tr>
<tr>
<td>$\beta$</td>
<td>Modal propagation constant</td>
</tr>
<tr>
<td>$\Delta$</td>
<td>Phase mismatch</td>
</tr>
<tr>
<td>$\Delta N_{eff}$</td>
<td>Effective index birefringence</td>
</tr>
<tr>
<td>$\Delta N_g$</td>
<td>Group index birefringence</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Relative electric permittivity</td>
</tr>
<tr>
<td>$\eta$</td>
<td>Efficiency</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>Coupling coefficient</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>Wavelength</td>
</tr>
<tr>
<td>$\Lambda$</td>
<td>Grating period</td>
</tr>
<tr>
<td>$\tau_{pulse}$</td>
<td>Temporal width of an optical pulse</td>
</tr>
<tr>
<td>$\chi^{(n)}$</td>
<td>Optical susceptibility of order $n$</td>
</tr>
<tr>
<td>$\omega$</td>
<td>Angular frequency</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

1.1 Background

Optical devices are commonly employed in different aspects of our daily lives. Application examples span from pollution sensing, LIDAR (light detection and ranging) and bio-imaging to automation, lightning and metrology, material processing and data communications over optical fibers. This has spurred continuous new developments, breakthroughs and innovation in optical materials, devices and systems.

Photonic Integrated Circuits (PICs) are systems combining multiple optical and functionalities devices on the same chip. In the quest for ever higher speeds and lower power consumption also for optical components, PICs have consequently attracted increasing interest during the past decades for applications ranging from biosensing and lab-on-a-chip systems [1, 2], to optical communications [3, 4] and quantum computing [5, 6].

Of all possible material candidates, many researchers have considered silicon (Si) as a well-suited template for PICs [7]. Si is the main element used in electrical integrated circuits and thus benefits from a well-established and mature fabrication technology. Si nanotechnology foundries have thus been adapted to accommodate also the needs of photonic devices, which can easily be integrated with the electronic devices on the same platform. Several optical devices such as ring resonators [8], low-loss optical waveguides [9, 10], Bragg gratings and delay lines [11, 12], variable attenuators [7], grating couplers [13] and switches [11, 14] have been realized over Si-based optical platforms, and in many cases have become commercially available [7]. However, optical systems realized on Si photonics platforms suffer from some crucial drawbacks. Since Si is an indirect-bandgap material, optical detectors and sources cannot be realized over this platform [15]. Furthermore, optical Si devices operating at optical telecommunication wavelengths require additional fabrication steps to suppress losses induced by two-photon absorption [16, 17]. Moreover, the lack of linear electro-optical properties in
unstrained Si implies that optical modulators in Si platforms need to be realized through other (generally slow) techniques, based on changing the concentration of free carriers [18], thermo-optics [19] or opto-mechanical effects [20]. Finally, the centrosymmetric crystalline structure of Si implies also the absence of a second-order parametric optical nonlinearity, $\chi^{(2)}$, in this platform. Consequently, nonlinear optical interactions in Si have to rely on, generally much weaker third-order effects ($\chi^{(3)}$ nonlinearity) [21].

The aforementioned drawbacks, on the other hand, do not apply for III-V semiconductors. Their direct energy bandgap allows detection and generation of optical signals with wavelengths covering the full telecommunication spectrum. Furthermore, their non-centrosymmetric crystalline structure results in intrinsic electro- and second-order nonlinear optical properties. These semiconductors have thus been employed for the realization of different PIC elements such as lasers and detectors, modulators and wavelength converters [22-25]. The effective exploitation of $\chi^{(2)}$ nonlinearities requires nevertheless advanced technological solutions, such as quasi-phase matching (QPM) [26], in order to counteract the deleterious effect of phase-velocity mismatch among waves at different frequencies, typically occurring due to material and waveguide dispersion. QPM schemes in III-V optical devices often involve complex technological processes involving the growth of orientation-patterned templates which increases propagation losses [27-29]. Furthermore, the electro-optic coefficients of III-V semiconductors are still quite low [30] with respect to oxide materials such as LiNbO$_3$, implying a relatively poor performance of this platform for electro-optical modulation.

Lithium Niobate (LiNbO$_3$, LN) is another template material employed for realization of optical devices. LN is a ferroelectric crystal with a wide transparency spectrum (0.4-5 $\mu$m wavelength range), simultaneously possessing piezo- and pyro-electric properties, combined with high electro- and nonlinear-optical features [31]. Furthermore, the development of a now well-established technology for QPM with periodically poled LN (PPLN) [32, 33], where ferroelectric domains are periodically inverted, has resulted in the widespread employment of LN for efficient nonlinear interactions [34, 35]. Thanks to its intrinsic high electro-optic coefficients ($r_{33} = 30$ pm/V) [31], high-end electro-optic modulators for e.g. ultrafast telecom systems in LN have been extensively studied [36-39] and are commercially available [40].

LN has also long been the material offering the most mature waveguide technology for integrated optics among all crystals used for electro-optics and QPM. Wave-guiding structures have traditionally been realized by ion-indiffusion or exchange methods such as proton-exchange [35, 37, 41-43] and Ti-diffusion [44, 45]. These methods yield refractive index contrasts typically in the range of 0.01-0.03 for optimal performance at telecom wavelengths. Accordingly, such waveguides have to be rather wide (>8 $\mu$m) and cannot afford sharp bending radii [46]. Even if combined with further processing to e.g. make ridge waveguides by etching [47-49] or dicing [50, 51], the ion-indiffused integrated devices in LN cannot possess small footprints. 
1.1. BACKGROUND

comparable to those achievable in silicon photonics, despite the relatively high refractive index of LN (~2.2).

Recently, a number of different approaches have been pursued to overcome the above mentioned drawbacks of traditional LN waveguides. An example consists in placing films or flakes of LN over nanophotonic PICs prepatterned in other templates, such as Si, to combine the benefits of high confinement waveguides with the nonlinear- and electro-optic properties of LN [52-56]. In such a case, however, a small portion of the power of the guided optical mode is actually confined in the LN films, which limits the achievable efficiencies of the electro- and nonlinear-optical interactions in such devices with respect to the ideal case of all-LN nanophotonic counterparts. Furthermore, a reliable and reproducible bonding of LN films over prefabricated devices induces additional complex technical steps. Such considerations have motivated several researchers to pursue an approach similar to smart-cut™ technologies for Si-on-insulator (SOI) platforms, that is bonding thin LN films ion-sliced from bulk crystalline substrates, via a low-index dielectric to a carrier substrate [57-59]. The use of the low index dielectric, typically silica or a polymer such as BCB, allows to realize a template providing a high index contrast. Such technology developments have most recently resulted in the commercial availability (essentially since 2015) of new thin film LN on Insulator (LNOI) substrates, attracting increasing interest as templates for the realization of miniaturized LN integrated optical devices. The relatively high refractive index contrast (0.7) offered by LNOI, enables guidance in submicrometer-sized optical devices, hence possessing much smaller footprints [46] and higher nonlinear- and/or electro-optical efficiencies than ion-diffused counterparts. Since 2015, an exponential growth of publications on the fabrication of LNOI-based PIC components, such as waveguides [60-63], ring and racetrack resonators [64-66], micro-disks [67-69] and grating couplers [70-75], and most recent demonstrations of ultrafast modulators and switches [76-78], wavelength converters [61, 69, 79-81], and Bragg filters [70, 71, 82] in LNOI nanophotonic waveguides, testifies to the interest and potential of this technology. Lastly, the very first periodically poled LNOI waveguides have also been reported [79, 83-85], paving the way towards ultracompact and efficient nonlinear interactions, in devices whose tight field confinements might theoretically boost efficiencies by two order of magnitude compared to current ‘large area’ PPLN waveguides.

The fabrication process for almost all aforementioned devices relies on dry etching, which implies significant technical challenges, given that LN is an inert material and cannot be easily etched. The employment of traditional fluorine-based dry etching methods generally results in redeposition of LiF [86] which in turn reduces the etching rates of LN and increases the roughness, and hence the propagation losses of the final device. Addressing such problems required consequent efforts to develop and refine a reliable fabrication process providing low-loss LNOI waveguides. This thesis is part of an intense research effort in this direction, pursued by a few groups at
international level, mostly in the US (Harvard, UCSB, CREOL), in the last two years. In this context is also to be mentioned the remarkable achievement by the Lončar group at Harvard in 2017 of a record low propagation loss of 2.7 dB/m [65].

As concluding remark on the appeal of LN in general and LNOI templates, in particular one can mention its interest for applications beyond (nano)photonic. This is related to its ferro-electric properties, and in particular its high spontaneous polarization ($P_s = 78 \mu\text{C/cm}^2$ [87]) and good-insulating properties, due to a large bandgap of $E_g = 3.78 \text{eV}$ [88] and high crystal quality, with extremely low defect-density. This implies significant amounts of bound charges at LN surfaces, with a huge and so far, still minimally unexplored potential for the local control of surface reactivity, stabilization [89] and molecular adsorption phenomena [90]. The fact that charge distributions at LN surfaces can be further engineered through PPLN structures opens up a brand new range of possibilities, with a huge and still largely unexplored potential for creating hybrid nanoscale-patterned organic-inorganic architectures, ferroelectric lithography [91], artificial photosynthesis [92] and a broad range of interdisciplinary applications, bridging photonics, ferroelectric science, biology and chemistry for sensing and manipulation from micrometric to nanometric scales. One example is biological sensing and spectroscopy with plasmon-enhanced photoluminescence and Surface Enhanced Raman Spectroscopy (SERS) on LN ferroelectric templates [93-97]. Other examples include particle and cell manipulation on biocompatible uncoated LN surfaces [98], via ferro-electric, photo-electric or acoustic effects [99-102].

1.2 Objectives and main achievements

The objective of this thesis was to take the LN technology and expertise of our group at KTH (on buried and ridge proton-exchanged waveguides and periodic poling in bulk crystals) and combine it with the expertise on Si-photonic I had acquired through my previous work at CREOL (Integrated Photonic Emerging Solutions group, led by Prof. S. Fathpour), with the goal of pushing the LN platform towards higher levels of integration and functionality by means of nanotechnology.

The ultimate vision is to achieve an integrated PIC platform of nanophotonic devices that can efficiently exploit the many appealing features of LN in formats compatible with those of silicon. Nanophotonic devices in LNOI are finally coming of age, also thanks to the efforts of a number of leading Si-photonic groups at international level which have entered the field in the last couple of years [65, 79, 103, 104]. In this context, this thesis marks a few important steps towards the achievement of this LN-photonic vision, with the development of a technology for in-house nanostructuring of LN and the achievement of a few novel experimental device demonstrators with it.
1.2. OBJECTIVES AND MAIN ACHIEVEMENTS

Reaching these objectives meant learning about LN material and waveguides, with their specific properties and needs for fabrication and characterization, mastering and developing nanofabrication and imaging techniques in the NanoFab Lab at AlbaNova and then pushing them to the levels enabling new devices. This required developing not only fabrication processes but also designs and theoretical/numerical analysis tools for my devices.

The first technological solution used in my thesis, also summarized in Paper A, involved fabricating sub-micrometric LN nanopillars (NPs) by Focused Ion-Beam milling, to exploit the tight confinement of optical modes in these devices and provide extra features in Second Harmonic Generation (SHG). Following studies on other platforms investigating the role of longitudinal field components in nonlinear nanophotonic waveguides [105, 106], this thesis demonstrates how they can be used to substantially modify and control the SHG polarimetric response of LN, with experiments coupled to comprehensive numerical analyses of the SHG interactions in LN NPs.

The FIB solution is not appropriate to develop a chip-scale planar PIC. For this reason, subsequent work in this thesis (since 2016) turned to developing nanopatterning techniques (Electron-Beam Lithography and etching) suitable for larger-scale processing on the first LNOI substrates, which at the time had just become commercially available. By adapting, developing and optimizing fabrication processes typical of Si-photonics to the LN platform, I could realize several LN-based PIC elements through a single fabrication pipeline. This fabrication process led to the demonstration of Bragg grating optical filters with record high aspect ratios (up to 45 dB at telecom wavelengths), comparable to the values reported in SOI and SiN platforms [12, 107]. The study, reported in Paper C, includes also one of the very first reported attempts for in- and out-coupling of optical signals to/from LNOI waveguides through grating couplers, with coupling losses below 10 dB. The experimental work, both at the design and at the characterization stages, was systematically backed by simulations of the PIC response. Ultimately a very good agreement between theory and experiments was achieved for all the PIC components reported in Paper C.

The next challenge was to further optimize the PIC fabrication process to reduce the device insertion losses. The progress made throughout this thesis, through the combination of fabrication and design efforts, has finally resulted in a reduction of the propagation losses in single-mode LNOI waveguides at telecom wavelengths, from the original 76 dB/cm [71] to the current 1.13 dB/cm.

Further results on the simulation side show also how the choice of waveguide geometry and cladding can be combined with the intrinsic properties of LN to design birefringence-free LNOI waveguides operating with fundamental modes only. The interest of these results, summarized in Paper B, lies in the fact that the intrinsic LN birefringence can relax fabrication tolerances and can also enable simultaneous group and phase velocity
matching in these devices, a feature not highlighted for any other nanophotonic platform so far.

Finally, another aspect investigated in this thesis in the framework of long-lasting collaborations of our group with scientists at the Conway Institute of Biomolecular and Biomedical Science, concerns novel interdisciplinary applications for LN devices to the life sciences. This thesis shows how LNOI surfaces, functionalized with silver nanoparticles, are an appealing template for Raman studies of biological specimens deposited on them (Paper D). LNOI templates yield a substantial enhancement in Raman signals due to their high index contrast, combined with engineered optical interference in the LN-SiO₂-Si multilayers and enhanced scattering obtained by etching (and in this specific case roughness is a desirable property). This thesis will also present how realization of topographical structures on LN, as well as engineered surface charges of PPLN devices, can be employed to control growth of neurons (Paper E).

1.3 Structure of the thesis

Chapter 2 will contain a brief review over LN properties. Description of dielectric properties of LN will be followed by representation of electro- and nonlinear-optical characteristics of this material. This chapter will furthermore provide a review over ferroelectric properties of LN.

In Chapter 3 theory of guided modes will be briefly discussed, explaining some physical characteristics implied by waveguiding phenomenon. This chapter will then briefly present theory of waveguide gratings, more detailed discussions of which are provided in Appendix A. Lastly, Chapter 3 will explain how electro- and nonlinear-optical interactions are affected by waveguide nature of integrated devices.

Different fabrication processes and characterization methods employed during this thesis are described in Chapter 4. Functionalities of each fabrication procedure, as well as the technical difficulties attributed to it, are explained in this chapter. Characterization methods employed to investigate fabricated LN structures are also represented in this chapter, including the topographical and optical characterization methods. When possible, I have also explained how each method can be employed to measure performance characteristics of fabricated optical structures.

Chapter 5 includes a summary of main results achieved during this thesis. The performance analysis of nonlinear processes occurring in LN nanopillars (Paper A), design of birefringence-free LNOI waveguides (Paper B), fabrication and characterization of high aspect ratio LNOI-based Bragg gratings (Paper C), and reduction of propagation losses inside fabricated LNOI waveguides are presented in this chapter. Some interdisciplinary applications of LN template, including employment of LNOI structures for enhancement of Raman signals (Paper D) and exploitation of topographical
and charged structures in LN surface for controlling cell growth dynamics (Paper E) are also described in Chapter 5.

Finally, Chapter 6 will summarize the major results achieved during this thesis. Based on simulation and fabrication evidences, I will also propose some potential directions for the future research exploiting functionalities of LN platform in PICs. Potential applications of such PIC elements for further interdisciplinary applications will also be presented in Chapter 6.
Chapter 2

Properties of lithium niobate

As discussed in Chapter 1, LN can be employed to realize many optical devices, some of which are even commercially available. There are also further functionalities associated with this material which arise from its crystalline structure and dielectric properties. This chapter will briefly describe the intrinsic properties of LN and explain how they can be used for different optical, but also electrochemical and biological applications.

2.1 Crystalline structure

Lithium niobate is an artificial ferroelectric material, transparent at wavelengths between 0.4 and 5 µm, and with relatively large nonlinear optical (NLO) and electro-optic (EO) coefficients [31]. High quality optical-grade LN wafers of congruent composition can be grown by the Czochralski technique [94] with low defect density, excellent stability and uniformity, and are commercially available in wafer diameters up to 5 inches (although the most common format is still 3 inches).

A schematic illustration of the LN crystalline structure is shown in Figure 2.1(a). Nb\(^{5+}\) and Li\(^+\) ions are placed between planes (orthogonal to the Z axis) where the O\(^{2-}\) ions sit. At temperatures below the Curie temperature (\(T_c \sim 1210°C\) [31]), the Li\(^+\) and Nb\(^{5+}\) ions are displaced with respect to the nearest O\(^{2-}\) plane and the middle of two planes, respectively, as shown in Figure 2.1(b). This provides a spontaneous polarization to the crystal, \(P_s\), oriented along the Z axis. The crystal is then in its ferroelectric phase, with a spontaneous polarization \(P_s = 78 \mu C/mm^2\) [87], which can be reversed by an electric field higher than the coercive field (\(E_c = 21 KV/mm\) [87]).

The Z crystalline axis of LN is defined as the one about which crystal exhibits a three-fold symmetry. LN possesses three equivalent axes (X) in its hexagonal unit cell, 120° apart from each other. The X axes lie in a plane
perpendicular to the Z axis and are perpendicular to the mirror symmetry planes, (Y, Z) planes, also with three-fold symmetry (see Figure 2.1(c)).

![Fig 2.1](image)

**Figure 2.1.** (a) Schematic representation of the LN crystal structure; (b) the displacement of the Li\(^+\) and Nb\(^{5+}\) ions with respect to the O\(^2-\) planes induces a spontaneous polarization, \(P_s\), along the Z axis; (c) schematic representation of the three-fold symmetry of LN: the mirror-symmetry planes (Y-Z axes) are indicated by solid blue planes. Subset indicates a possible combination of (X, Y, Z) axes with respect to symmetry planes (solid blue lines).

### 2.2 Dielectric permittivity

If principal dielectric axes of a material are the same as reference coordinate system, the electric flux density, \(D\), and electric field, \(E\), inside dielectric will be related as:

\[
D = \begin{pmatrix} D_x \\ D_y \\ D_z \end{pmatrix} = \varepsilon_0 \varepsilon_r E = \varepsilon_0 \varepsilon_r \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \varepsilon_0 \begin{pmatrix} \varepsilon_{11} & 0 & 0 \\ 0 & \varepsilon_{22} & 0 \\ 0 & 0 & \varepsilon_{33} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}
\]

(2.1)

where \(\varepsilon_0\) is the electric permittivity of vacuum and \(\varepsilon_r\) is the relative dielectric permittivity tensor of the material (rank 2), which has –generally– complex elements. The real and imaginary parts of these elements are related to material dispersion and losses, respectively. Since the relative dielectric permittivity matrix is diagonal in the principal dielectric axes reference frame, one can replace \(\varepsilon_{11}, \varepsilon_{22}\) and \(\varepsilon_{33}\), by \(\varepsilon_X, \varepsilon_Y\) and \(\varepsilon_Z\), respectively, where the indices 1, 2 and 3 refer to the axes X, Y and Z, respectively.

LN is symmetric about its Z axis, hence its permittivity is the same for any electric field polarization perpendicular to it, and thus \(\varepsilon_X = \varepsilon_Y\), meaning that LN is a uniaxial material, and Z is its optic axis. At low frequencies (i.e., from zero up to radio frequencies), the values of the LN relative permittivity are \(\varepsilon_X = \varepsilon_Y = 84\) and \(\varepsilon_Z = 30\) [31].
At optical frequencies the transparency spectrum of LN is rather wide (ranging from 0.4 µm to 5 µm). The relative permittivity is then real and directly related to the refractive index of the material, \( n \), as \( \varepsilon_{r,i} = n_i^2 \), where index \( i = X, Y, Z \) denotes the crystalline axis.

### 2.2.1 Refractive index

At optical frequencies, the refractive index of a material changes as a function of wavelength (and temperature) and can be approximated by Sellmeier equations of the form:

\[
n^2 = A_1 + \frac{A_2 + B_1 F}{\lambda^2 - (A_3 + B_2 F)^2} + B_3 F - A_4 \lambda^2
\]

where \( n \) and \( \lambda \) are the refractive index of the material and the wavelength (in units of µm) of the electromagnetic wave and \( A_i \) and \( B_i \) are material- (and eventually also polarization-) dependent constants. The term \( F \) expresses the temperature dependence and for LN can be written as \( F = (T - T_0)(T + T_0 + 546) \), where \( T_0 = 24.5 \) °C is a constant and \( T \) is the temperature (in units of °C) \([108]\). The wavelength-dependence of a material’s refractive index, described by the Sellmeier equation, is called material dispersion.

**Figure 2.2.** Ordinary (solid blue curve) and extraordinary (dashed red curve) refractive indices of LN at room temperature, as a function of wavelength.

Since the refractive index of a material is directly related to its relative dielectric permittivity and LN is uniaxial with \( Z \) as its optic axis, its refractive indices depend also on the field polarization and can be expressed as \( n_X = n_Y = n_o \) and \( n_Z = n_e \), where \( n_o \) and \( n_e \) are called ordinary and extraordinary refractive indices of the material, respectively. Sellmeier equations of the form of are then given separately for \( n_o \) and \( n_e \) with suitable \( A_i \) and \( B_i \) coefficients \([108]\). Figure 2.2 illustrates the material dispersion of
LN at room temperature, both for the ordinary and extraordinary refractive indices. As it can be seen, the former, $n_o$, is larger than the latter, $n_e$, which means that LN is negatively birefringent.

The index ellipsoid of LiNbO$_3$

The anisotropy of optical materials (eq. 2.1) is often described also through a geometric representation, the so-called *index ellipsoid*. This can be introduced by considering the energy density of the electric field stored inside the medium, $u_e$, given by:

$$u_e = \frac{1}{2} \mathbf{D} \cdot \mathbf{E} = \frac{1}{2} \varepsilon_0 (\varepsilon_x E_x^2 + \varepsilon_y E_y^2 + \varepsilon_z E_z^2)$$ \hspace{1cm} 2.3

By defining $x = D_x/(2 \ u_e \varepsilon_0)^{1/2}$ (and similarly for $y$ and $z$), and substituting $\varepsilon_i$ by $n_i^2$, eq. 2.3 can then be rewritten, in the following form:

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1$$ \hspace{1cm} 2.4

which represents an ellipsoid (the index ellipsoid) in three-dimensional space. Eq. 2.4 holds for the case where principal dielectric axes of the material are the same as the reference coordinate system, i.e.: $(x, y, z) = (X, Y, Z)$. Otherwise, the off-diagonal elements in the permittivity tensor given in eq. 2.1 (and hence the refractive index matrix $n^2=\varepsilon_r$) are non-zero. When material symmetries are considered, $\varepsilon_{r,ij} = \varepsilon_{r,ji} = n_{ij}^2 = n_{ji}^2$ and reduced Voigt notation can be used, substituting the two indices $i$ and $j$, for one index, $h$, [109]:

$$h = \begin{cases} 
1 & \text{for } ij = 11 = XX \\
2 & \text{for } ij = 22 = YY \\
3 & \text{for } ij = 33 = ZZ \\
4 & \text{for } ij = 23 \text{ or } 32 \\
5 & \text{for } ij = 13 \text{ or } 31 \\
6 & \text{for } ij = 12 \text{ or } 21 
\end{cases} \hspace{1cm} 2.5$$

Then (when the reference frame does not coincide with the crystal principal dielectric axes) the index ellipsoid takes the general form given by:

$$\left(\frac{1}{n_i^2}\right) x^2 + \left(\frac{1}{n_2^2}\right) y^2 + \left(\frac{1}{n_3^2}\right) z^2 + \left(\frac{1}{n_4^2}\right) yz + \left(\frac{1}{n_5^2}\right) xz + \left(\frac{1}{n_6^2}\right) xy = 1$$ \hspace{1cm} 2.6
2.3 Electro-optics

The linear electro-optic (EO) effect, also called as Pockels effect, implies a linear change of the refractive index of a material induced by a static (or low frequency) electric field \([109]\). This effect can be employed to realize devices such as tunable waveplates, electrically-controlled optical modulators, and tunable filters and resonators. The EO effect is present in crystals lacking a center of symmetry, such as III-V semiconductors and ferroelectrics.

The change in refractive index or rather of each component of the dielectric permittivity matrix in the Vogt notation \((\varepsilon_h = n_h^2; h = 1, 2, \ldots 6)\) is related to the vectorial components of the static electric field, \(E_k (k = 1, 2, 3\) represent \(X, Y, Z,\) respectively), as according to \([109]\):

\[
\Delta \left( \frac{1}{n_h^2} \right) = \sum_k r_{hk} E_k
\]

where \(\Delta (1/n_h^2)\) denotes the change in refractive indices (or dielectric permittivity tensor) and \(r_{hk}\) represents the EO tensor of the material in the Voigt notation (reducing the 3\(^{rd}\) rank EO tensor, \(r_{ijk}\), to a 6×3 matrix, \(r_{hk}\)).

The 6×3 matrix of EO coefficients for LN is given by:

\[
r_{hk} = \begin{pmatrix}
0 & -r_{22} & r_{13} \\
0 & r_{22} & r_{13} \\
0 & 0 & r_{33} \\
r_{42} & 0 & 0 \\
r_{42} & 0 & 0 \\
-r_{22} & 0 & 0 \\
\end{pmatrix}
\]

where \(r_{22}, r_{13}, r_{33}\) and \(r_{42}\) are 6.7, 10.9, 34 and 32 pm/V, respectively \([31]\).

Applying an electric field to an EO material can in general change its index ellipsoid from the simple form of eq. 2.4 to the form of eq. 2.6, meaning that the off-diagonal elements in the refractive index matrix of the material can become nonzero. In order to calculate the changes of the refractive indices induced by EO effect, one needs to find a new reference frame in which the permittivity tensor takes a diagonal form (readers are referred to \([110]\) for more details).

As a simple (and practical) case, suppose that an electric field is applied to the LN crystal, along its \(Z\) crystalline axis (\(E = E_z \hat{z}\)). In such a case, \(\Delta (1/n_0^2) = \Delta (1/n_2^2) = r_{13} E_z\) and \(\Delta (1/n_3^2) = r_{33} E_z\). The new index ellipsoid is thus expressed as:

\[
\left( \frac{1}{n_0^2} + r_{13} E_z \right) x^2 + \left( \frac{1}{n_0^2} + r_{13} E_z \right) y^2 + \left( \frac{1}{n_3^2} + r_{33} E_z \right) z^2 = 1
\]

Assuming small changes in the refractive index, one can use a Taylor series expansion to obtain the new values of the refractive index:
\[ n'_{X} = n'_{Y} \approx n_0 - \frac{1}{2} n_0^3 r_{13} E_z \]  
\[ n'_{Z} \approx n_e - \frac{1}{2} n_e^3 r_{33} E_z \]  

The choice of this specific example is made deliberately here, since in most of the cases one wants to exploit the highest EO coefficient of LN \( (r_{33}) \), by applying a field along \( Z \) to modify the refractive index of light polarized along the same axis, according to eq. 2.11. The employment of electro-optical effects in waveguide-based devices will be treated in next chapter.

### 2.4 Nonlinear optics

Applying an electric filed, \( E \), to any optical medium results in an induced polarization, \( P \) [109], which can be phenomenologically expressed as:

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

where \( \chi^{(i)} \) stands for the \( i \)th order optical susceptibility tensor (of rank \( i+1 \)) of the medium. The first term in eq. 2.12 describes linear effects and is directly linked to the permittivity tensor, i.e. \( \varepsilon_r = 1 + \chi^{(1)} \). The higher order terms on the right-hand side of eq. 2.12 constitute the nonlinear polarization of the medium. The \( \chi^{(i)} \) susceptibility generally decreases as higher orders of \( i \) are considered. Furthermore, materials with a center of symmetry have zero \( \chi^{(i)} \) susceptibilities for even values of \( i \).

LN is non-centrosymmetric and hence it has a nonzero second order nonlinearity tensor \( \chi^{(2)} \), with relatively high values. Thus, in what follows we shall only on the second term in eq. 2.12, leading to the second-order optical nonlinearity: \( P^{(2)} = \varepsilon_0 \chi^{(2)}EE \). Furthermore, we shall use Voigt notation to reduce the \( \chi^{(2)} \) tensor into 3×6 elements, which we –according to experimental practice– express with nonlinearity matrix, \( d \), with coefficients given by: \( d_{ih} = \frac{1}{2} \chi_{ih}^{(2)} \), with \( i=1, 2, 3 \) and \( h=1, 2, \ldots 6 \), similarly to eq. 2.5 [111].

\[
\begin{pmatrix}
P_X \\
P_Y \\
P_Z
\end{pmatrix}_{SH} = \varepsilon_0 \begin{pmatrix}
d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\
d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\
d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36}
\end{pmatrix}
\begin{pmatrix}
E_X^2 \\
E_Y^2 \\
E_Z^2 \\
2E_Y E_Z \\
2E_Z E_X \\
2E_X E_Y
\end{pmatrix}_{FF}
\]
2.5. FERROELECTRICITY

In case of LiNbO$_3$, the second-order nonlinearity matrix, $d$, has the following form:

$$d = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$$

where $d_{31} = 4.8$ pm/V, $d_{22} = 2.2$ pm/V and $d_{33} = 25.7$ pm/V [31].

2.5 Ferroelectricity

LN is an electric insulator which can be regarded as a high bandgap semiconductor ($E_g = 3.78$ eV [88]), and a ferroelectric material with a large spontaneous polarization ($P_s = 78$ µC/cm$^2$). The ability to locally invert the polarization, creating ferroelectric domain arrays in periodically poled LN (PPLN) structures is extensively used in nonlinear optics for quasi phase-matching (QPM) purposes, as mentioned briefly in next chapter.

From an electrostatic point of view, the PPLN technology allows also to create bound charge distributions at ferroelectric interfaces defined by the ferroelectric-nonferroelectric interfaces (see Figure 2.3). The low defect density and insulating properties of LN imply that such surface charges will be externally screened. Thus, LN surfaces can be extremely active in attracting ions, charged molecules and particles.

![Figure 2.3](image)

**Figure 2.3.** Schematic representation of domains with opposite polarities in a PPLN structure. Positive (green) and negative (red) bound charges at ferroelectric interfaces (dashed circles) are also depicted.

The active LN surfaces can be controlled by ferroelectric domains, to yield engineered electrochemical state and adsorption characteristics [112, 113], electron affinity [114] and surface potential [115] at PPLN surfaces. Interesting effects can also be observed at the atomically thin boundaries where polarity changes in PPLN structures, called as domain walls (shown schematically in Figure 2.3), possessing enhanced localized electric fields [94] and in some cases also different electrical properties [116] than single-domain bulk LN crystals. Such properties, do not only depend on the polarization of LN ferroelectric domains, but can be additionally controlled by the normal tools used and developed to make waveguides and pattern micro- and nano-structures at LN and LNOI surfaces, including wet and dry etching, masking
and other cleanroom processing steps [96, 115, 117]. These aspects and potential additional control parameters provided by the LN nanotechnology toolbox were also partially explored in this thesis, through international collaborations of our group with scientists in Ireland and in the USA. In the framework of the former, this thesis includes also two devices I made for quite different and interdisciplinary applications to life science, which will be briefly explained here.

2.5.1 LN templates for cell growth patterning

The biocompatibility of uncoated LN surfaces can be combined with its ferroelectric patterning capabilities (e.g., in PPLN structures) to control growth of biological specimens such as cells. It has been shown that the polarity of LN domains can affect proliferation of stem cells [99, 118]. Ferroelectric lithography of biological species, moreover, is viable by exploiting domain walls. Domain walls have not only been shown to control directionality of axons in neuron culturing [98], but have also been employed for photochemical deposition of nanostructures comprising organic molecules [119].

2.5.2 LN templates for photodeposition

Experiments over different liquids such as water, methanol and acetic acid reveal that the surface polarization of LN domains affects its absorption characteristics and surface chemistry [117, 120]. The polarization of ferroelectric domains can also be manipulated to control the local electronic structure and chemical reactivity on LN surfaces, allowing for novel fabrication of complex structures [121]. Advanced patterning of LN surfaces with the rich fabrication toolkits developed for integrated and nonlinear optics in this material, including periodic poling, ion-exchange, etching and additional treatments[95-97, 122], and be used to engineer local photoreduction processes and drive selective photodeposition of metallic nanoparticles and nanoparticle arrays in a controllable way.

2.5.3 LN templates for Raman enhancement

Raman scattering, also known as Raman effect, is the inelastic scattering of a photon by molecules, through which the molecules are excited (relaxed) to different vibrational modes. As a consequence, the energy of the photons scattered by the molecule decreases or increases by very specific amounts [123]. Each molecule leaves its unique fingerprints in the spectrum of its Raman scattered light, allowing its (label-free) identification and even a quantification of its abundance. Accordingly, Raman spectroscopy (spectral
analysis of the back-scattered Raman signal) is employed in a broad range of applications such as geology [124], semiconductor surface analysis [125] and life sciences [126].

Raman interactions are, however, extremely weak, and a lot of schemes to enhance their signals have been explored and developed in the past two decades. Surface-Enhanced Raman Spectroscopy (SERS) employs plasmonic effects on nanopatterned noble metal surfaces, where localized metal features act as nano-antennas whose plasmonic resonances, can dramatically enhance the interaction of light with the Raman-scattering species in the proximity of these ‘hot-spots’ [127]. One critical feature is the need to controllable fabricate metal patterns with very small features ~10nm to maximize these effects. This is often at the limit of current top-down approaches and drives the interests into alternative, bottom up approaches, using, e.g. self-assembled metal nanoparticles. The bound charge (and electrostatic field distributions) on ferroelectric patterned LN templates have been proven to drive the successful photodeposition and arrangement of silver [96, 97] and gold [122, 128] nanoparticles from aqueous solutions under UV illuminations. Such photodeposited metallic nanoparticles, chemically patterned over LN domains, have been then exploited in SERS technology to efficiently detect biological particles and even single molecules [129, 130].

---

a A noble metal is a metal resistant to corrosion and oxidation.
Chapter 3

Theory of guided wave devices

This chapter contains a basic theory of guided wave devices, in addition to a brief explanation of how waveguidance affects the response of LN optical devices, when compared to their bulk counterparts. It provides a short introduction to optical waveguide theory and to the concept of guided modes and of their effective and group indices, transverse and longitudinal field components, as well as waveguide dispersion and birefringence. Grating-assisted guided-mode interactions and electro- and nonlinear-optical effects are also discussed in the framework of coupled-mode theory in waveguide configurations.

For the sake of generality, the equations of this chapter are written in a generic cartesian system with \((x, y, z)\) axes, related to the waveguide geometry and to be distinguished from the principal dielectric axes of LN, \((X, Y, Z)\), introduced in Chapter 2. The relative orientation of the two reference frames may vary depending on the envisaged device and application and shall be duly specified in relation to the latter whenever necessary (e.g. in Chapter 5).

3.1 Optical waveguides

Optical waveguides (herein called simply as waveguides) are dielectric structures in which optical signals propagate without experiencing diffraction. A waveguide is generally formed by a core material with a refractive index higher than the surrounding (substrate and cladding). Light is confined in the regions of higher refractive index in the transverse plane, here denoted as \((x, y)\) and propagates in the orthogonal (longitudinal) direction, here given by the axis \(z\). Figure 3.1 illustrates some common waveguide structures. In all structures, the refractive index of the core, \(n_c\), is higher than those of substrate \((n_s)\) and cladding \((n_d)\). The cross-sectional dimensions of waveguides in the \((x, y)\) plane may span from \(\sim1\ \mu\text{m}^2\) up to \(\sim100\ \mu\text{m}^2\), depending on the
wavelength of the optical signal, the materials and the geometry used for the waveguide. In the next section the main equations concerning waveguides and some of their properties employed in this thesis are briefly reviewed.

Figure 3.1. Some commonly used geometries to build optical waveguides: (a) circular, (b) slab, and (c) ridge waveguides. z is the propagation direction.

3.1.1 Maxwell’s equations and guided modes

In a lossless, nonmagnetic and dispersionless medium with no free charges, Maxwell’s equations can be written as [30]:

\[ \nabla \cdot \mathbf{D} = 0 \]  

\[ \nabla \cdot \mathbf{B} = 0 \]  

\[ \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \]  

\[ \nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t} \]

where \( \mathbf{D} \) and \( \mathbf{E} \) are the electric flux density and electric field vectors, respectively (related through the polarization expressed by eq. 2.1 in the linear case). \( \mathbf{B} \) and \( \mathbf{H} \) are the magnetic flux density and magnetic field intensity vectors, respectively, which are related through the magnetic permeability of vacuum, \( \mu_0 \), as: \( \mathbf{B} = \mu_0 \mathbf{H} \), inside a nonmagnetic medium.

From the Maxwell’s equations (eqs. 3.1-3.4) with some additional steps and approximations, one can obtain the wave equation:

\[ \nabla^2 \mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2 \mathbf{E}}{\partial t^2} = 0 \]  

where \( n = n(x, y) \) is the refractive index distribution of a (weakly guiding) waveguide in its transverse plane, and \( c = 1/\sqrt{\varepsilon_0 \mu_0} \) is the propagation speed of light in vacuum.

For light at an angular frequency of \( \omega = 2\pi c/\lambda \), propagating along the +z direction with a propagation constant of \( \beta \) the electric field can be written as:
3.1. OPTICAL WAVEGUIDES

\[ \mathbf{E}(x, y, z; t) = \Re \{ \mathbf{E}(x, y)e^{j(\omega t - \beta z)} \} \]  

where \( \mathbf{E}(x, y) \), the electric field transverse distribution, is a vector, which in general can have transverse (along \( x \) and \( y \)) and longitudinal (along \( z \)) components with complex values.

By substituting eq. 3.6 into the wave equation and defining transverse Laplacian as \( \nabla_T^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \), eq. 3.5 can be recast into the Helmholtz equation:

\[ \nabla_T^2 \mathbf{E} + \left( \frac{\omega^2}{c^2} n^2 - \beta^2 \right) \mathbf{E} = 0 \]

Substituting for \( n^2 \) with the local refractive indices of the materials used for building the waveguide in the \((x, y)\)-plane (e.g. those schematically presented in Figure 3.1) and using the boundary conditions for the fields, the Helmholtz equation can be solved. For a waveguide, this yields an eigenvalue problem, with a discrete set of solutions, which are called optical guided modes. Such solutions are characterized by one or two integers (mode indices), in the case of waveguide with one-dimensional (Figure 3.1(b)) or two-dimensional (Figure 3.1(a) and (c)) confinement, respectively. The \( \mu \)-th order mode, denoted by the generic mode index \( \mu \) and propagation constant \( \beta_\mu \) and field profiles \( \mathbf{E}_\mu(x, y) \) and \( \mathbf{H}_\mu(x, y) \), the total electric field being then given by:

\[ \mathbf{E}_\mu(x, y, z; t) = \Re \{ \mathbf{E}_\mu(x, y)e^{j(\omega t - \beta_\mu z)} \} \]

The eigenvalues \( \beta_\mu \) have a lower bound given by the propagation constants in the lower index (substrate and/or cladding) material and upper bound given by the propagation constant in the core:

\[ \frac{\omega}{c} \max(n_c, n_s) < \beta_\mu < \frac{\omega}{c} n_c \]

The power flow of a guided optical mode with index \( \mu \), carried along the propagation direction (\( z \)), can be expressed in terms of the transverse vectorial components of the electric and magnetic fields of the guided mode, \( \mathbf{E}_{\mu} \) and \( \mathbf{H}_{\mu} \), respectively, as [131]:

\[ P_\mu = P_{z,\mu} = \Re \left\{ \frac{1}{2} \int \int \left[ \mathbf{E}_{\mu} \times \mathbf{H}_{\mu}^* \right] dx \, dy \right\} \]

\[ = \frac{1}{4} \int \int \left[ \mathbf{E}_{\mu} \times \mathbf{H}_{\mu}^* + \mathbf{E}_{\mu}^* \times \mathbf{H}_{\mu} \right] dx \, dy \]

At a given wavelength, the guided modes of a waveguide form a complete orthonormal set, i.e., for two generic guided modes \( \mu \) and \( \nu \), with amplitudes normalized for unitary power flow (\( P_\mu = 1 \) in eq. 3.10), is written as:

\[ \frac{1}{4} \int \int \left[ \mathbf{E}_{\mu} \times \mathbf{H}_{\nu}^* + \mathbf{E}_{\nu}^* \times \mathbf{H}_{\mu} \right] dx \, dy = \delta(\beta_\mu - \beta_\nu) \]

where \( \beta_\mu \) and \( \beta_\nu \) stand for the propagation constants of the two guided modes.
Longitudinal components of the guided modes

Decomposing the $\mathbf{E}$- and $\mathbf{H}$-fields of the generic guided modes of index $\nu$ into their transverse ($\mathbf{E}_{\nu T}$ and $\mathbf{H}_{\nu T}$) and longitudinal ($\mathbf{E}_{\nu L}$ and $\mathbf{H}_{\nu L}$) components, and using Maxwell’s equations, one can obtain the following relations [131]:

\[
\mathbf{E}_{\nu L} = -j \frac{1}{\varepsilon \omega} \nabla_T \times \mathbf{H}_{\nu T} \quad \text{(3.12)}
\]

\[
\mathbf{H}_{\nu L} = j \frac{1}{\mu \omega} \nabla_T \times \mathbf{E}_{\nu T} \quad \text{(3.13)}
\]

Eqs. 3.12-3.13 imply that the tight spatial confinement (high gradients of the transverse field distributions) of the guided modes in e.g. nanowaveguides may yield non-negligible longitudinal ($z$-polarized) components of the electromagnetic waves trapped in the waveguide, at variance from usual (purely Transverse Electro-Magnetic (TEM)) waves in free-space.

Transverse Electric (TE) and Transverse Magnetic (TM) modes

The wave equation for a slab waveguide (Figure 3.1(b)), admits one further degree of freedom, associated to the field polarization. One finds that the electric field for some guided modes is perpendicular to the plane of incidence (the plane containing propagation direction, $z$, and the unit vector normal to the interfaces of optical materials forming waveguide, $y$, in Figure 3.1(b)), i.e., $\mathbf{E} = \mathbf{E}_x$, while the magnetic field has $\mathbf{H}_y$ and $\mathbf{H}_z$ components. Such modes are called Transverse-Electric (TE) [131]. On the other hand, in the case of Transverse-Magnetic (TM) modes, the magnetic field is perpendicular to the plane of incidence, and hence the mode only contains $\mathbf{H}_x$, $\mathbf{E}_y$ and $\mathbf{E}_z$ components.

In the case of two-dimensional confinement as in ridge waveguides (Figure 3.1(c)), the $\mathbf{E}$-fields ($\mathbf{H}$-fields) of all guided modes contain both $\mathbf{E}_x$ and $\mathbf{E}_y$ ($\mathbf{H}_x$ and $\mathbf{H}_y$) components, and pure TE (TM) modes do not exist. Nonetheless, the guided modes may have dominant electric or magnetic fields perpendicular to the plane of incidence, and thus are called quasi-TE or quasi-TM. These modes are commonly labelled by two integers $m$ and $n$, where $m$ ($n$) represents the number of zeros in the transverse profile of the guided mode along the horizontal (vertical) axis. For the sake of simplicity, hereafter quasi-TE$_{mn}$ (quasi-TM$_{mn}$) modes are simply called TE$_{mn}$ (TM$_{mn}$) modes.

Effective refractive index and group index

For each guided mode of index $\mu$ (dropped in the next equations) at a given frequency $\omega$, an effective refractive index, $N_{\text{eff}}$, can be defined, according to the following relation:
\[ \beta = \frac{\omega}{c} N_{\text{eff}} = \frac{2\pi}{\lambda} N_{\text{eff}} \]

\( N_{\text{eff}} \) is the refractive index the guided mode “feels” while propagating in the waveguide, and hence the velocity of propagation of a monochromatic signal in the waveguide, that is its phase velocity, can be written as:

\[ v_p = \frac{\omega}{\beta} = \frac{c}{N_{\text{eff}}} \]

and the guidance condition presented in eq. 3.9 yields:

\[ \max(n_{\text{cl}}, n_s) < N_{\text{eff}} < n_c \]

When dealing with optical pulses (i.e. not monochromatic signals around a carrier frequency \( \omega_0 \)), an additional quantity is introduced to characterize the propagation speed of the pulse, namely the group velocity. In the case of a guided-mode of propagation constant \( \beta \), this is given by [30, 132]:

\[ v_g = \frac{\partial \omega}{\partial \beta} |_{\omega_0} = v_p + \lambda_0 \frac{\partial v_p}{\partial \lambda} |_{\lambda_0} \]

Accordingly, a mode group index can also be defined, given by:

\[ N_g = \frac{c}{v_g} = \frac{\partial \beta}{\partial \lambda} |_{\lambda_0} = N_{\text{eff}} - \lambda_0 \frac{\partial N_{\text{eff}}}{\partial \lambda} |_{\lambda_0} \]

### 3.1.2 Waveguide dispersion

The effective index of a guided mode depends on the materials and geometry used to build the waveguide, and on the wavelength of the optical signal. The chromatic dependence of \( N_{\text{eff}} \), and hence of the propagation constant \( \beta \), arising from the waveguide properties (and not from the dispersion of the materials used to form the waveguide) is called waveguide dispersion. The dispersion behavior of a waveguide can be engineered by a proper choice of waveguide geometry [133-137] and materials employed for its construction [133, 134, 138].

As an example, the simulation results shown in Figure 3.2 illustrate the waveguide dispersion for the first two guided TE and TM modes (TE\textsubscript{00} and TM\textsubscript{00}, respectively), for a 1 \( \mu \)m-wide and 300 nm-thick waveguide in thin film Lithium Niobate on Insulator (LNOI) with no cladding layer (in other words, the cladding is simply air). The dashed black line shows the refractive index of \( \text{SiO}_2 \) (substrate material), indicating the cut-off condition for the mode guidance (see eq. 3.16). The effective indices in Figure 3.2 were computed by COMSOL Multiphysics, a commercially available software based on Finite-Element Method (FEM) solving the vectorial wave equation in the waveguide.
3.1.3 Waveguide birefringence

The refractive index experienced by an optical signal when propagating in a birefringent medium depends on the light polarization. In contrast to bulk materials, where birefringence is an intrinsic property of the medium (see section 2.2.1), most rectangular waveguides possess (or form) waveguide birefringence, i.e., different effective indices of the TE and TM modes (as an example, see Figure 3.2).

Although birefringence of a waveguide can have some applications such as in polarization maintaining fibers [139], it also induces polarization-dependent phase shifts and time delays, which can in turn negatively affect efficiencies, extinction ratios and bandwidths of switching and modulating PIC devices [134, 140]. This issue and methods to eliminate it will be addressed in section 5.2.

3.1.4 Coupled-mode equations in waveguide configurations

The discussion so far concerned a waveguide with linear polarization $P_0$, related to the non-homogeneous index distribution which defines the waveguide, $n(x, y)$, by the relationship $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}_0 = \varepsilon_0 (1 + \chi^{(1)}) \mathbf{E} = \varepsilon_0 n^2 \mathbf{E}$. Periodic modulations of a waveguide structure, electro-optic effects or alternatively nonlinear optical effects, if weak, can be treated as small
perturbations to the original waveguide structure defined by $P_0$ (the unperturbed waveguide) whose modal structure was defined in the previous sections. The perturbation, in all of the cases listed above, can be described by the addition of a perturbative term $\Delta P$ to the original polarization $P_0$ of the waveguide, so the total polarization is now $P = P_0 + \Delta P$.

When $\Delta P \neq 0$, the modes of the perturbed system are no longer the same in the new system, i.e. they interact and exchange energy. If $|\Delta P| \ll P_0$, nevertheless, a perturbative approach can be taken, assuming that the original waveguide modes are slightly perturbed, in the sense that their amplitude may change upon propagation, due to the interaction. This effect is described by looking for new solutions of the form $A_\mu(z)E_\mu(x,y)e^{j(\omega t - \beta_\mu z)}$ where the amplitude of each (original) mode $\mu$ is modulated by an envelope function $A(z)$, slowly varying in propagation ($z$). The evolution problem is then reduced to analysis the coupling of the original modes and deriving the set of coupled-mode equations governing the evolution along $z$, for the –complex– envelopes of the modes $A_\mu(z)$, with a procedure detailed in [30, 131].

### 3.2 Waveguide gratings

*Waveguide gratings* are periodic modulations of the waveguide properties with periods comparable to the optical wavelength [131]. They can be implemented by locally varying the refractive index in the waveguide core, substrate or cladding materials, but also by periodically changing the waveguide dimensions. Figure 3.3 schematically shows examples of waveguide gratings on slab waveguides, which may be used to: (a) couple co-propagating or (b) counter-propagating guided modes (of orders $\mu$ and $\nu$ in the figure) or (c) couple a guided-mode $\mu$ to a radiation mode, all at the same frequency. Waveguide gratings are widely used in applications such as wavelength dispersion engineering [137, 141] and reflection of optical signals [71]. As it will be described in Appendix A, these waveguide gratings can be employed to reflect some guided optical modes. In this case, the gratings are called *distributed Bragg reflector*.

Consider a grating with a period of $\Lambda$, implemented by periodically changing the relative dielectric permittivity of the waveguide media along the propagation direction $z$. The perturbation in the dielectric permittivity, $\Delta \varepsilon(z)$, can be expanded in a Fourier series [131]:

$$\Delta \varepsilon(z) = \sum_q \Delta \varepsilon_q(\kappa)e^{-jqKz} = \sum_q \Delta \varepsilon_q(\kappa)e^{-jqK} \quad 3.19$$
where \( K \) is the grating vector, parallel to grating propagation direction \((z)\), and has a magnitude of \( K = |K| = 2\pi/\Lambda \). Now, consider two guided modes, \( \mu \) and \( \nu \), propagating in the waveguide with wave vectors of \( \beta_\mu \) and \( \beta_\nu \), respectively, which experience the grating modulation. Using the coupled-mode formalism \([131]\), the interaction with the two modes induced by the grating can be described through evolution equations for the slowly varying amplitudes, \( A_\mu(z) \) and \( A_\nu(z) \), respectively, of these two modes, written as follows \([131]\):

\[
\pm \frac{d}{dz} A_\mu(z) = -j\kappa^* A_\nu(z) e^{-j2\Delta z}
\]

\[
\pm \frac{d}{dz} A_\nu(z) = -j\kappa A_\mu(z) e^{+j2\Delta z}
\]

The \( \pm \) sign on the left-hand sides of eqs. 3.20-3.21 are defined by the propagation directions of the two modes (modes propagating in the \((-z)\) direction have a negative sign). Eqs. 3.20-3.21 contain two key parameters. One is the phase mismatch, defined as:

\[
2\Delta = \beta_\mu - (\beta_\nu + qK)
\]

where \( q \) is the order of coupling relevant for the grating. The corresponding \((q^{th})\) coupling coefficient between modes \( \mu \) and \( \nu \) is the other key parameter, given by:

\[
\kappa = \kappa_{\mu\nu}^{(q)} = \frac{\omega\varepsilon_0}{4} \int E_\mu^*(x,y)\Delta\varepsilon_q(\kappa)e^{-jqKz}E_\nu(x,y) \, dx \, dy
\]

where \( \kappa_{\mu\nu}^{(q)} \) is simply written as \( \kappa \). If we consider a case where \( \beta_\mu = -\beta_\nu \) (i.e., mode \( \nu \) is the reflection of mode \( \mu \) caused by the grating), one can solve the coupled mode equations to obtain the reflection of optical signals caused by the Bragg grating as \([131]\):

\[
R = \frac{|A_\nu(0)|^2}{|A_\mu(0)|^2} = \left( 1 + \frac{1 - \Delta^2/\kappa^2}{\sinh^2 \left( L\sqrt{\kappa^2 - \Delta^2} \right)} \right)^{-1}
\]
A more detailed treatment of Bragg gratings structures using coupled-mode equations and Transfer Matrix Methods (suitable also for the strong perturbation cases) is provided in Appendix A.

### 3.2.1 Phase matching for grating couplers

Figure 3.3(c) illustrates coupling of a radiation mode into a guided mode via a grating coupler. The same structure can also function oppositely (i.e., can couple a guided mode into a radiation mode). If a radiation mode with propagation constant of \( n_{cl}\beta_0 \) (\( \beta_0 \) being its propagation constant in vacuum) is radiated into the grating coupler structure with an angle of \( \theta \) as shown in Figure 3.3(c), and its component along waveguide direction (\( z \)) is denoted as \( \beta_q \), it can be coupled into (or out of) the waveguide structure if phase matching condition given in eq. 3.25 is satisfied [131]:

\[
\beta_q = n_{cl}\beta_0 \sin \theta = \beta_\mu + qK; \quad q = 0, \pm 1, \pm 2, \ldots
\]

where \( q \) is the coupling order. Given the wave-guiding condition of eq. 3.16, the phase-matched coupling occurs only if \( q = -1 \) [131]. Figure 3.3(c) shows the vectorial phase matching conditions for a grating coupler in such a case.

### 3.3 Electro-optics in waveguides

As discussed in section 2.3, the linear electro-optic effect can be used to change the refractive index of an electro-optical medium. The latter, in turn, will result in a change of the effective refractive indices of the guided modes inside a waveguide. Figure 3.4(a) provides an example of how two electrodes with a spacing of \( G \) can be employed to apply an electric field to an LN waveguide.

However, the electro-optic relations in waveguides slightly differ from those in bulk medium. In such a case, the overlap between the guided mode inside the waveguide and the electric field induced by an external voltage, \( V \), needs to be considered. Furthermore, one needs to deliberate the use of different materials in the waveguide, where in most cases substrate and cladding materials do not have electro-optical properties. As an example, if we consider the structure presented in Figure 3.4(a), the change in the effective refractive index for a guided \( TE_{00} \) mode will be [36, 40]:

\[
\Delta n(V) = -\frac{1}{2} N_{eff}^3 \frac{V}{G} \Gamma
\]

\[
\Gamma = \frac{G}{V} \iint E(x,y)r(x,y)E_{opt}(x,y) \cdot E_{opt}^*(x,y) \, dx \, dy
\]
where $J$ is the overlap between the applied electric field, $E$, and the transverse distribution of the guided optical mode, $E_{\text{opt}}$ (the “opt” indexing is used here to distinguish the electric field $E$ from the optical mode, $E_{\text{opt}}$). It is also assumed that the guided mode is normalized (based on eq. 3.10) to have a power flow of unity. In eq. 3.28, the electro-optic coefficient $r$ is written as $r(x,y)$, accounting for the fact that its value changes for different material used to build the waveguide.

EO modulation is probably the most recognized example application of EO effect in waveguides. LN, very well-known for its intrinsic electro-optical properties, is widely used to realize EO modulators with waveguide structures. Although LN-based EO modulators are nowadays commercially available, there are still several research groups trying to exploit and optimize EO operation of LN waveguides. Reduction of propagation and coupling losses, as well as improvements in bandwidths of such devices, have significantly improved performance characteristics of LN-based EO modulators [40]. Although employment of indiffusion techniques to realize waveguiding structures in LN results in low-loss devices, EO modulators based on such devices suffer from high driving voltages and relatively large footprints. In contrast, the high index contrast offered by LNOI platform enables researchers to realize submicrometric waveguides allowing for smaller distance between EO electrodes, which in turn results in a decreased driving voltage in such devices.

### 3.4 Nonlinear optics in waveguides

This section explains how nonlinear interactions briefly described in section 02.4 need to be modified in a waveguide geometry. Suppose that an optical mode $\mu$ with a frequency of $\omega_{\text{FF}}$ propagates along $z$ inside a nonlinear optical media, as schematically depicted in Figure 3.5.
Eq. 2.13 suggests that a second-harmonic of this mode can be generated inside the nonlinear waveguide with a frequency of $\omega_{SH} = 2\omega_{FF}$, coupled into a guided optical mode $v$. The propagation of unperturbed optical modes $\mu$ and $v$ inside the waveguide, along $z$ axis, can be described as:

\[ E_\mu(x, y, z) = A_\mu(z) F(x, y) e^{-j\beta_\mu z} \]  \hspace{1cm} 3.28

\[ E_v(x, y, z) = A_v(z) G(x, y) e^{-j\beta_v z} \]  \hspace{1cm} 3.29

where $F(x,y)$ and $G(x,y)$ stand for transverse distributions of the guided optical modes $\mu$ and $v$, respectively, and $A_i(z)$ denotes the slowly varying envelope of each mode. However, presence of the nonlinear media results in a perturbative term in the Helmholtz equation for each guided mode [111, 142]:

\[ \nabla_T^2 E + \left( \frac{\omega^2}{c^2} n^2 - \beta^2 \right) E = -\mu_0 \omega^2 P^{(NL)} \]  \hspace{1cm} 3.30

where the term $\partial^2/\partial t^2$ in Helmholtz equation is replaced by $-\omega^2$, and $P^{(NL)}$ is the second-order nonlinear polarization. Eq. 2.13 describes $P^{(NL)}$ for the mode $v$, and can be rewritten as [111]:

\[ P_v^{(2)} = \varepsilon_0 d_{eff} E_\mu E_\mu \]  \hspace{1cm} 3.31

where $d_{eff}$ is an effective value of $d_{ih}$ nonlinearity matrix presented in eq. 2.13. A detailed derivation of $d_{eff}$ can be found in [111].

In such a case, the amplitudes of the modes $\mu$ and $v$ are related through coupled mode equations for the nonlinear interaction [142]:

\[ \frac{\partial A_v}{\partial z} = -j\kappa_v A_\mu^2 e^{j\Delta \beta_{SHG} z} \]  \hspace{1cm} 3.32

\[ \frac{\partial A_\mu}{\partial z} = -j\kappa_\mu A_v A_v^* e^{-j\Delta \beta_{SHG} z} \]  \hspace{1cm} 3.33

where propagation losses of the optical modes are neglected, $\Delta \beta_{SHG} = \beta_v - 2\beta_\mu$ is the phase-mismatch for the Second-Harmonic Generation (SHG) process, and:

\[ \kappa_v = \frac{\omega_{SH}}{\pi c N_v} \cdot \frac{\iint d_{eff} E_\mu^2(x, y) E_v(x, y) dx dy}{\iint E_v^2(x, y) dx dy} \]  \hspace{1cm} 3.34
\[ \kappa_{\mu} = \frac{2\omega_{FF}}{\pi c N_{\mu}} \cdot \frac{\iint d_{eff} E_{\mu}^2(x,y)E_{\nu}(x,y)dx\,dy}{\iint E_{\mu}^2(x,y)dx\,dy} \]  \hspace{1cm} 3.35

If we assume an undepleted pump regime (i.e., if the conversion efficiency is low and pump power after a propagation length of \( L \) is approximately similar to the input pump power), the conversion efficiency for the SHG process can be written as:

\[ \eta_{SHG} = \frac{P_{SH}(L)}{P_{FF}(0)} \]
\[ = \frac{8d_{eff}^2}{\varepsilon_0 c N_{eff,SH} N_{eff,FF}^2 \lambda_{SH}^2 S_{eff}} \sin^2 \left( \frac{\Delta \beta_{SHG} L}{2} \right) L^2 P_{FF,0} \]  \hspace{1cm} 3.36

where \( S_{eff} \) stands for effective cross section of the nonlinear interaction, and is expressed as:

\[ S_{eff} = \frac{(\iint E_{FF}^2(x,y)dx\,dy)^2 \iint E_{SH}^2(x,y)dx\,dy}{(\iint E_{FF}^2(x,y)E_{SH}(x,y)dx\,dy)^2} \]  \hspace{1cm} 3.37

The SHG conversion efficiency, \( \eta_{SHG} \), described in eq. 3.36, changes as a \( \sin^2 \) function of \( L \), suggesting periodic generation and depletion of the SH signal. However, if one can achieve a \textit{phase-matched} case, where \( \Delta \beta_{SHG} = 0 \), \( \eta_{SHG} \) will be quadratically depend on \( L \). \textit{Modal phase matching} is one solution for the phase mismatch issue. In such a case, the waveguide is designed such that the interacting modes \( \mu \) and \( \nu \) feel the same effective refractive index \([81]\). Moreover, existence of gratings, by means of changing waveguide width \([81]\) or by periodically inverting ferroelectric domains of the LN \([79, 143]\), can also lead to quasi phase matching of the SHG process.
Chapter 4

Fabrication and characterization methods

This chapter will provide an overview of different fabrication processes used in this thesis and resulting in the devices investigated and used for Papers A-E. It presents the main technical challenges to be overcome to attain the desired devices performances. The last part of the chapter introduces briefly the main techniques used for structural and optical characterizations of the fabricated devices.

4.1 Fabrication methods

Figure 4.1 presents an overview of the processes employed and developed in this thesis. The flow-charts show the fabrication steps relevant for each process. In general, the main hurdle to be overcome when patterning devices in LiNbO₃ is its resilience to etching, as confirmed by extensive attempts performed in the field over the years, also prior to this work [86, 144-146]. This is a particularly critical aspect when aiming at nanophotonic devices involving submicrometric waveguides and gratings, such as the ones of papers A-C, generally requiring etching depths of 0.3-1 μm, as well as smooth sidewalls and fine features (down to ~150nm), with good reproducibility and uniformity, in order to guarantee the desired device functionalities and minimize insertion losses. It is very difficult, if not impossible, to use a single fabrication method to meet all such demanding requirements. Therefore, the approach followed in this thesis was to explore different fabrication techniques and choose and optimize ad-hoc fabrication recipes for each target device and application.

The nonlinear optical experiments in LN nanopillar waveguides (Paper A) required a fine control of the individual pillar diameter (with resolutions down to ~25 nm), smooth and vertical sidewalls and etching depths of ~1 μm. To this aim, the process of choice was focused ion-beam (FIB) milling,
performed according to the steps outlined in Figure 4.1(a). When carefully optimized, FIB milling resulted in pillars with sizes ranging from 125 to 350 nm, with ultra-smooth and almost vertical (85°) sidewalls. The FIB process is discussed in section 4.1.1.

![Figure 4.1](image)

**Figure 4.1.** Flow-charts for the different fabrication processes employed in this thesis. (a) Nanopillar fabrication by focused ion beam milling; (b) waveguide fabrication on LNOI by electron-beam lithography and etching; (c) periodic poling of bulk LN. Pictures: (a) scanning electron microscopy image of a fabricated nanopillar, (b) atomic force microscopy image of a fabricated waveguide grating, (c) piezo-response force microscopy image of two-dimensional hexagonal ferroelectric domains in a Z-cut PPLN crystal.

However, the FIB process is very time-consuming and cannot be used for large area device fabrication. Therefore, a completely different approach, involving electron-beam lithography (EBL) and subsequent LN etching via a hard mask, was developed to fabricate the integrated devices considered in **Paper C**, requiring chip-scale (several mm-long) sub-micrometric photonic ridges. This process was then expanded with further device functionalization involving planar side-electrodes for electro-optic tuning, periodic structures for fiber-to-chip coupling and spectral filtering, and additional cladding layers for loss minimization. This is illustrated by the diagram in Figure 4.1(b) and discussed in section 4.1.2.

The technology outlined in Figure 4.1(b) was used also for the design and implementation of the devices of **Papers D-E** (with standard photolithography), to pursue novel applications of LN to Raman spectroscopy.
and cell growth in controlled microfluidic environments, in collaboration with researchers at University College Dublin and Conway Institute of Biomolecular and Biomedical Research (Dublin).

Finally, the last ingredient of the technology toolbox used in this thesis involves the fabrication of periodically poled devices in bulk samples, which were used for further investigations of cell growth on PPLN (Paper E) and studies of ferroelectric domain stability and switching ([96], not included in the thesis). The fabrication steps involved in the poling process are listed in Figure 4.1(c) and described in more details in section 4.1.3.

### 4.1.1 Focused ion beam milling

The structures of Paper A were realized using the SEM/FIB dual beam system (FEI Nova 200) at the AlbaNova Nano-Fabrication Laboratory. The FIB milling process used a Gallium ion beam generated through a 30 kV filament, and I have performed several ad-hoc attempts to optimize the fabrication process enabling us to reproducibly realize nanopillars with smooth and vertical sidewalls. The final optimized process employs a thin chromium layer uniformly deposited on the LN chips to avoid any charging effects (particularly pronounced and deleterious for LN, given its excellent electrical insulating properties) during the milling process. The other parameters involved in the FIB process, namely the scan modality (raster vs. serpentine), the dwell time and the ion beam current, were thoroughly optimized to finally obtain a reproducible milling process. A low ion beam current was found to be important to guarantee extremely small roughness, which nevertheless resulted in relatively long milling times (~25 minutes) for each nanopillar. The scanning modality of the ion beam and its dwell time were found also to be important factors to realize high quality nanopillars with vertical sidewalls. Examples of how different parameters can affect the FIB milling results are illustrated in Figure 4.2. Comparing Figure 4.2 (a) and (b), one can see that raster-type scanning results in nanopillars with more vertical sidewalls. Furthermore, a comparison between Figure 4.2 (c) and (d) suggests that prolonging the dwell time from 1 µs to 5 µs allows to make longer nanopillars.

Ad-hoc optimization of the FIB process resulted in the reliable production of nanopillars in all the different cuts (X-, Y- and Z-) of bulk LN and also on thin film LNOI substrates, for nanopillar sizes down to 125 nm, heights of ~1 µm, with very smooth and almost vertical (~85°) sidewalls.
Figure 4.2. Examples of how different parameters of the ion beam can affect the quality of LN nanopillars fabricated by FIB milling: (a) serpentine vs. (b) raster scan, and raster scan with (c) 1 μs and (d) 5 μs dwell times. Nanopillars made with an ion beam current of 30 pA. Scale bars represent a length of 1 μm.

4.1.2 Technology of waveguide fabrication

Although the fabrication of photonic devices by FIB milling can yield extremely smooth sidewalls, this method is not well-suited for the realization of integrated optical circuits. It requires long fabrication times for each device and loses its resolution when aiming for larger write fields, and thus cannot be used to fabricate long (millimeter-scale) submicrometric waveguides, nor fine and long periodic features such as Bragg gratings. As a result, I decided to take a different approach, enabling the chip-scale functional devices of Paper C, based on the development of electron-beam lithography (EBL) and dry etching techniques on LNOI. The process was also used with standard photolithography (instead of EBL) whenever fabrication tolerances could be relaxed (>2 μm), as was the case of the devices of papers D-E.

Here, the implementation of the full device fabrication pipeline, concerning the advance patterning of sub-micrometric waveguides in LNOI, with the ancillary steps of electrode patterning and waveguide cladding deposition, will be described. In its final optimized implementation, this process enabled the production of multiple PIC functional elements (gratings, waveguides, ring resonators, Bragg reflectors, tapers) at the chip scale through the same fabrication procedure, outlined by the flow chart of Figure 4.1(b), using a single lithographic step. As it will be explained in this section, extra care had to be taken in all fabrication steps in order to minimize the roughness of the final LNOI structures. In fact, nanophotonic waveguides impose very stringent demands on the tolerable roughness of the fabricated structures, since imperfection and inhomogeneity results in scattering of the optical fields, which will in turn translates into increased optical losses.
The workflow for the final fabrication process of waveguide-based LN optical devices is shown in Figure 4.3. It consists of: a) dicing, b) deposition of a Chromium (Cr) layer on LN, c) spin-coating and patterning of resist through lithography, d) a first dry etching step to transfer the pattern from the resist to the Cr, e) a second dry etching step to transfer the pattern from the Cr to the LN layer, and, finally, f) removal of the remaining Cr from the fabricated LN device. Each step is briefly explained in the following.

**Figure 4.3.** Fabrication steps in the realization of LNOI integrated optical devices: (a) diced thin-film LNOI-on-silicon chip, (b) Chromium hard mask deposition, (c) spin-coating and soft-baking of a resist layer, (d) lithography and pattern-transfer by dry etching of Cr, (e) dry etching of LN, (f) removal of the Cr mask. The arrows indicate the crystalline axes of the LN thin film. \( w_0 \) is the width of the fabricated LN feature (e.g. waveguide).

### Thin-film LNOI wafers

The process starts with commercial LNOI (Papers C, D) wafers, but can be equally applied to bulk LN crystals (Paper E). In the case of paper C, the original wafer consists of a 3-inch 300 nm-thick Y-cut LN layer bonded through a ~2 \( \mu \)m-thick SiO\(_2\) layer to a 4-inch carrier Si wafer.

### Cr deposition

A uniform Chromium (Cr) layer is first deposited on the LNOI chips using an electron gun deposition system (Edwards Auto 306 FL400). This Cr layer acts as a hard mask for the subsequent dry etching of LN. Cr was chosen based on previous studies [86, 144, 145, 147] and previous work in our group [94, 147], because of its relatively high selectivity in the dry etching of LN. Furthermore, the Cr serves as a conductive layer below the resist film during the EBL step to minimize charging effects during exposure.
Lithography for patterning LN devices

The desired patterns to be ultimately etched in LN are transferred into the chromium hard mask through a lithographic process. Lithography is the most commonly used step to fabricate micro/nano structures on different substrates. The applications can range from single components for research purposes, to commercial fully integrated circuits, transistors, etc. Depending on their desired feature sizes, the devices of this thesis have been fabricated by either optical or electron-beam lithography. The former was suitable for reliably etching feature sizes in waveguide devices down to 2 µm (Papers D-E), while the latter was used whenever higher resolutions (<100 nm) were required (Paper C).

Optical lithography (photolithography)

The optical lithography (or photo-lithography) process is based on the optical illumination of a photosensitive film (photoresist), through a predesigned photomask containing the desired pattern. The photoresist is typically spread onto the sample surface through spin-coating, followed by a soft baking step. The photoresist layer is then exposed to a wavelength within its sensitivity spectrum. According to the Rayleigh criterion the resolution of an optical system scales linearly with the wavelength [148], and hence standard sources for photolithography are generally within the ultraviolet (UV) spectrum. Mercury lamps are frequently used for this purpose, with emission wavelengths at 436 nm (g-line), 406 nm (h-line) and 365 nm (i-line).

During exposure, a photomask is placed between the optical system (delivering illumination to the sample) and the sample to be exposed. After a suitable exposure time (depending on the type of photoresist, its treatment conditions, the wavelength and power of the source, and the exposure pattern), the resist is immersed in a chemical developer solution to reveal the mask patterns. A positive resist leads to the removal of the exposed areas, while in the case of a negative resist, the unexposed areas will be dissolved by the developer. I routinely used a positive photoresist (S1813, Microposit®), providing film thicknesses of 1.3 and 1.2 µm when spin-coated at speeds of 4000 rpm and 6000 rpm, respectively. Following spin-coating, soft-baking of the photoresist was performed by heating the sample in a convection oven (Memmert UP400). UV illumination was then performed by a Karl Suss mask aligner (MJB3), employing the g-line of a mercury lamp. After resist development (MF319, Shipley Microposit®), the photoresist was finally hard-baked to increase its thermal, chemical and physical stability in the subsequent processing steps.
4.1. FABRICATION METHODS

**Electron Beam Lithography (EBL)**

The resolution obtained by photolithographic methods may not be suitable for realization of optical devices with submicrometric feature sizes. Other lithography methods, such as electron beam lithography (EBL), which can generate feature sizes down to few tens of nanometers should be employed. An EBL system directly writes the desired patterns by exposing an *e-beam resist* to electron beams controlled by an internal pattern generator. Hence, it is a maskless process. The electron beam is generated by an electron gun, accelerated with energies in the range of few tens of kV, and passed through suitable apertures and alignment coils. A deflecting system is finally used to deflect the electron beam to address the desired points on the resist area. Figure 4.4 schematically shows the structure of an EBL system. Similar to the photolithographic method, the pattern on the e-beam resist is revealed by immersion in a suitable developer after e-beam exposure.

![Figure 4.4. Schematic representation of an EBL system. Inset: illustration of the results of exposure with a given e-beam pattern on a positive (top) and negative (down) resist (in grey).](image)

Because of the small area the deflection system can cover without losing electron beam quality, the pattern to be finally realized on the e-beam resist is
generally divided into smaller sub-patterns (called write fields). After exposure of each write field, the sample is moved by a positioning system so that the EBL system can continue to pattern the next write field. The mechanical movement of the sample under the e-beam for each write field can lead to misalignments between patterned structures within two adjacent write fields. These misalignments, called stitching errors, are to be avoided as they can critically degrade the optical performance of the fabricated devices. Misalignments occurring along an optical waveguide act as scattering center, which increase the propagation losses for the guided signals. Even more critical are stitching errors along periodic structures such as Bragg gratings, where even tiny misalignments can induce unwanted phase shifts which may seriously affect the optical performance.

Minimizing the stitching errors is crucial for optical devices such as waveguides, exhibiting lengths of a few millimeters (to be compared with typical write-fields of 100-500 µm). To overcome such an issue, whenever possible, I employed the Fixed-Beam Moving-stage (FBMS) capability of our EBL system (Raith Company, Turnkey 150), in which the sample is continuously moved under the e-beam when exposing the pattern. This method, however, can be applied only to path-like structures (and not to e.g. grating fabrication). The EBL system used in this thesis produced electron beams with an acceleration voltage up to 25 kV and a nominal write-field size of 100×100 µm². An aperture size of 10 µm was typically used for patterning fine structures, while an aperture size of 20 µm was used to pattern larger structures such as waveguide tapers, where resolutions are less critical for the correct performance of the optical device. The final optimized recipe for EBL patterning used a negative 300 nm thick e-beam resist (ma-N 2403, Micro-resist Technology®) revealed by ma-D 525 developer (Micro-resist Technology®).

Dry etching

The transfer of a desired pattern from the resist into substrate material, is generally performed by a selective etching of the surface of the latter, through the openings of a hard (etch-resistant) mask with the desired pattern. Typical etching processes can be divided into two categories: wet and dry etching. In the former case, a liquid-phase etchant is employed to remove the unmasked parts of the material. Given its chemical nature, the wet etching can provide a large etching selectivity between the mask material and the substrate, when the mask and etchant solution are suitably chosen. However, in many cases the wet etching is not completely anisotropic and cannot provide vertical sidewalls. Dry etching, on the other hand, involves the removal of material from the substrate by physical means, by bombarding it with ions that dislodge portions of the material from the exposed surface. The dry etching process is generally more anisotropic than wet etching, but this happens at the
expense of a reduced selectivity, due to the physical nature of the etching process.

In this thesis, Inductively Coupled Plasma Reactive-Ion Etching (ICP-RIE) was employed to transfer the pattern from resist films to Cr hard masks and then from the Cr masks into the LN thin films. During ICP-RIE, a plasma of desired ions is formed at low pressures by a magnetic field (ICP). A separate parallel-plate combination is simultaneously employed to create a directional electric field near the sample, to achieve more anisotropic etching profiles. The balance between physical and chemical etching can be adjusted through the choice of the applied RF and ICP powers, the etching gas mixture and pressure. Figure 4.5 schematically shows how an ICP-RIE system is built, as well as some typical isotropic/anisotropic etching profiles. In this thesis an Oxford Plasmalab 100 system was used. The ICP and RF generators of this system can be powered up to 500 W. The system can also be cooled down to temperatures down to $-70^\circ$C, using a flow of liquid nitrogen.

**Cr etching**

The first step to be optimized for a faithful transfer of the patterned features into the underlying LN layer, is the Cr etching. The goal here is to have an anisotropic etching (i.e., with vertical sidewalls as of in Figure 4.5(c)), with high selectivity of the Cr versus that of the resist film. The sidewall roughness of the etched Cr structures is another important factor to be considered, since any roughness in the sidewalls of the Cr hard masks is potentially transferred into the etched LN structures. In the final optimized recipe, I used a mixture of Cl$_2$ and O$_2$ gases and a temperature of 30°C. The resulting etch-rates for the Cr and resist films were $\sim$10 nm/min and $\sim$6-7 nm/min, respectively, yielding a selectivity of $\sim$1.5. An example of the resulting etched Cr hard mask is shown in Figure 4.6(a).
CHAPTER 4: FABRICATION AND CHARACTERIZATION METHODS

Figure 4.6. Some dry etching results: (a) etched hard mask in chromium. Etching results for LNOI waveguides made by photolithographic patterning: (b) LN dry-etched in SF$_6$/Ar, with evidence of LiF redeposition on the waveguide walls ((image taken before Cr removal), (c) LN dry-etched as in (b), but with an additional wet-etch step to remove the LiF layer. Results for EBL-patterned structures in LNOI: (d) waveguide etched in a SF$_6$/Ar gas mixture and (e) waveguide and sidewall grating etched by Ar-ion milling, and (f) smooth waveguide sidewalls obtained with the final an optimized Ar-ion milling recipe. Scale bars represent lengths of 1 µm.

**LN etching**

It is very difficult to pattern LN, a chemically inert material, by dry etching methods. Due to the high volatility of fluorinated niobium species, fluorine-based plasmas are generally employed for chemical dry etching of LN [86]. Several studies in the literature have been devoted to optimizing the etching conditions with fluorinated gases such as SF$_6$, CHF$_3$, and CF$_4$, mixed with an inert gas (generally Ar) [86, 144, 145]. However, chemical fluorine-based etching of LN leads to the formation of LiF, which in turn is redeposited on LN, reducing the overall etch rate. Furthermore, the redeposition of LiF leads to roughness on the sidewalls of the etched areas, as seen in Figure 4.6(b). There are nevertheless ways to alleviate these deleterious effects by adding further processing steps [63, 146], which in turn increase the time required to realize the final LN devices.
I first developed and optimized a dry etching recipe for LN devices. I used a gas mixture of SF$_6$ and Ar to chemically etch LN structures at a temperature of $-10^\circ$C, which resulted into structures with sidewall angles of $\sim 60^\circ$ (with respect to the horizontal axis), etched at a rate of $\sim 30$ nm/min, with a selectivity of $\sim 1.5$ with respect to the Cr hard mask.

Although extra care was taken in the processing, the fabricated devices still exhibited a rms roughness of $\sim 23$ nm on their sidewalls (Figure 4.6(b)). To reduce this effect, I implemented a new recipe with the approach of [146], where additional wet-etching steps are used to remove the LiF that gets deposited after the RIE process. For this purpose, the fabricated samples were immersed in a 2:2:1 volume mixture of NH$_4$OH:H$_2$O$_2$:H$_2$O at a temperature of 85°C, in three steps of one hour each. With this method I could reduce the rms sidewall roughness to 12.6 nm (Figure 4.6(c)), yet these results did not prove to be reproducible.

As an alternative solution, I then explored the option of pure physical etching of LN, by means of Ar$^+$-ion milling [61, 65, 71, 78, 80, 81, 149]. The LN structures etched by Ar-milling can have much smoother sidewalls than those etched by fluorine-based plasmas, which is extremely beneficial for reducing optical losses. Indeed, an exceptionally low propagation loss of 2.7 dB/m was recently reported by the Lončar group at Harvard, in SiO$_2$-cladded LNOI waveguides fabricated by Ar milling [65]. The sidewall angles of Ar-milled structures are, nonetheless, less steep than those obtained by fluorine-based etching. Furthermore, the etching rates of Ar-milled LN devices are generally lower than those of fluorine-based etching, resulting in longer processing times. The latter, however, is not too critical an issue considering the relatively limited thicknesses (usually less than 500 nm) to be etched in LNOI in order to produce tightly confining waveguides at 1550 nm, thanks to the high index contrast provided by LN versus SiO$_2$.

Ar-milling was therefore the method I finally chose to develop and optimize (since 2017) and which I used to realize the LNOI PIC devices presented in Paper C. As an example, Figure 4.6(e) shows a waveguide grating fabricated through Ar milling in an ICP-RIE machine. Besides granting the high structuring resolutions required for fine-pitch Bragg gratings ($\sim 250$ nm features), my final recipe resulted in an rms roughness of less than 5 nm. It involved Ar-milling at a pressure of 10 mTorr and a temperature of $-50^\circ$C. The etched devices possessed a sidewall angle of $40$-$45^\circ$, and were etched with a rate of 3-4 nm/min. As an example, Figure 4.6(f) shows an SEM image of a 300 nm-thick LN waveguide fabricated with the optimized recipe.

**Cr removal**

After etching the LN films to realize desired optical structures, the last step consists of removing the remaining Cr hard mask. Otherwise, the presence of any residual metallic structure on top of the waveguide causes significant
propagation losses of the optical modes. I have used a commercially available ceric ammonium nitrate-based etchant to etch away any remaining Cr mask (Sigma-Aldrich Co., 651826). Although the nominal etch rate of Cr in this etchant is >200 nm/min at room temperature\(^b\), the processed LNOI chip needed to be immersed in this etchant for longer times to ensure a full removal of all the metal.

**Electric contacts for Electro-Optics: lift-off**

An additional element in the LNOI toolbox, required in order to exploit EO effects in the fabricated LNOI waveguides, involves the deposition and patterning of electrical contacts at the sides of photonic wires, as schematically shown in Figure 4.1(a). For this purpose, I have developed a lift-off process and suitable alignment procedures for a second EBL patterning step. This time I used a positive e-beam resist, AR-P 6200.18 (Allresist Co.). After spin-coating of the resist and soft-baking inside a convection oven, EBL was performed with a dose slightly higher than that required for a normal lithography, followed by a development step longer than that specified in the datasheet, in order to realize resist sidewalls with negative angles (undercut) as schematically shown in Figure 4.7(a).

A metallic layer was then deposited on the samples by an electron-beam evaporation. First a thin (5-10 nm) Cr layer was deposited at a rate of 1-1.5 Å/s, which was followed by deposition of a gold (Au) layer with a thickness

---

\(^b\) Value provided by etchant datasheet.
comparable to that of the etched LN devices (see Figure 4.7(b)). The Cr layer serves as an adhesion promoter between the gold and the substrate. Finally, the chip was immersed and sonicated in acetone to remove the remaining resist and lift-off the metallic stripes sitting on top of it, to achieve the desired electrical contact patterns, as shown schematically in Figure 4.7(c).

PMMA cladding

For the last generation of my LNOI devices I used an additional PMMA cladding, serving as a passivation layer for the fabricated devices and protecting them from contaminations. Furthermore, in view of planned experiments based on electro-optical effects, the PPMA reduces the possibility of electric breakdown when voltages are applied across the waveguides and, having a relative dielectric permittivity higher than air, it helps confining more of the electric field inside the LN waveguide and thus increases the efficiency of the electro-optical device. Last but not least, the cladding layer can significantly reduce propagation losses of a waveguide structure [150].

4.1.3 Periodic poling of LiNbO₃

Arrays of ferroelectric domains of alternating polarity implemented by electric field poling in LN are key to maximizing the efficiency of frequency conversion devices in a broad wavelength range. Periodically poled LN (PPLN) is the key technology tool to realize quasi phase-matching, compensating for the phase mismatch faced by e.g. SHG process, as discussed in section 3.4. Moreover, in more recent years PPLN structures have proven to be useful also for novel applications such as cell growth, ferroelectric lithography and nanoparticle deposition.

The process for periodic poling of LN is schematically shown in Figure 4.1(c). Photolithography techniques are normally used to pattern an insulating photoresist layer on bulk Z-cut LN chips. Electrical contacts are made across the LN crystal through the opening in the resist on one side and the uncovered backside of the sample by means of a conductive electrolyte gel. The patterned electrodes define the regions of the samples where the applied external field is going to drive a local domain inversion. This is achieved by connecting the sample cell to an external poling circuit, sketched in Figure 4.8(a) designed and optimized by our group in KTH [94]. The poling setup includes a signal generator, providing the voltage waveform to be applied across the sample, the output of which goes through a high-voltage amplifier (which applies a constant gain, equal to 2000). The amplified waveform is then applied to a cell used for the poling, in which contacts are made to the conductive gel to transfer the voltage to the LN sample, as shown in Figure 4.8(b). Meanwhile, the voltage and current passing through the poling cell are monitored in real time. Figure 4.8(c) shows an example of the poling voltage
(blue curve) and current (green curve) recorded during the poling of a 0.5 mm-thick congruent LN sample.

**Figure 4.8.** (a) Schematic diagram of the poling electrical setup. (b) Sketch of poling configuration with electrical contacts on LN made through the openings of a patterned electrically-insulating resist layer. (c) Typical voltage and current waveforms for the periodic poling of a 0.5mm-thick congruent LN crystal in the self-termination regime.

Ferroelectric domain inversion begins as soon as the electric field applied to the LN sample exceeds the coercive field value, $E_c = 21$ KV/mm, at which point the LN sample (originally insulating) becomes conductive. The current passing through the poling cell increases and then, in the case of an optimal, self-terminating process, it spontaneously decreases when the total charge deposited on the sample (during the poling) reaches the value of $Q = 2 P_s A$, where $A$ is the surface area of the regions to be poled (openings in the photoresist) and $P_s = 78 \mu$C/cm$^2$ is the spontaneous polarization of LN. Self-termination can be seen in the current waveform shown in Figure 4.8(c), where the current drastically decreases by its own, even if the poling voltage is still applied (poling plateau).
4.2 Characterization methods

Several methods have been employed in this thesis to characterize topography, optical properties and performance of the fabricated devices. This section provides a quick overview on them.

4.2.1 Scanning electron microscopy

Scanning electron microscopy (SEM) is commonly used to analyze topographic features with much higher resolutions than optical microscopes. An SEM system has a structure similar to that of an EBL system, in which the electron beam scans the surface of the sample. The interaction of the electron beam with the sample surface produces scattered electron signals containing information about sample topography and composition and can be therefore used for imaging. The SEM images of this thesis were obtained with the system installed in the FIB milling machine (FEI Nova 200).

4.2.2 Scanning probe microscopy

Scanning Probe Microscopy (SPM) is another method used to characterize the properties of specimen at nanometric scales. The simplest common form of SPM used for surface topography imaging is Atomic Force Microscopy (AFM). An AFM system measures surface profiles by monitoring the deflection of a cantilever whose tip is kept in contact (or close to contact) with the surface of the specimen, while scanning over its surface. The deflections of the cantilever are detected by the differential output of a 2×2 detector array, recording the reflection of a laser beam from the backside of the cantilever. The resolution of the method is ultimately limited by the size of the AFM tip (typically ~10-20 nm). A basic AFM system can be further functionalized to implement more advanced SPM techniques, to sample locally not only topography, but also elastic, electric and ferroelectric properties at the surface of the sample.

Piezo-response Force Microscopy (PFM) is an advanced form of SPM, used to image domains in ferroelectric materials. It is based on the change of sign of the piezoelectric coefficients across ferroelectric domains of opposite polarity and uses the inverse piezoelectric effect to detect their polarity. A high frequency (compared to the rate of changes in surface topography) voltage signal is applied to the surface of material via a conductive AFM. The voltage applied to the ferroelectric (piezoelectric) material leads to a local expansion or compression of the material in phase or out of phase with the voltage, depending on the local sign of the piezoelectric coefficients. The sample deformation is picked up as an AFM signal by the cantilever in contact with the surface. The amplitude and phase of the mechanical oscillations of the cantilever at the frequency of the applied voltage yield information about the
polarity of the ferroelectric domains and the amplitude of the spontaneous polarization in the material, e.g. in a PPLN sample. Readers are referred to [151] for a more detailed explanation of the PFM method.

4.2.3 Optical characterization methods

The key features of optical (linear or nonlinear) devices are typically characterized in terms of their polarization response, spectral or angular tuning and efficiency (or losses). This applies also to integrated optical devices, where further experimental complexity and ad-hoc methods are required to couple light in and out of the device and evaluate its performance. This is briefly discussed in the following paragraphs, with specific reference to the evaluation of coupling and propagation losses in waveguide devices and polarimetric studies of their nonlinear response.

**Butt-coupling**

Butt-coupling is a well-established method to couple light into waveguides. This is achieved by focusing the input radiation (from free space or an optical fiber) on the polished end-facet of a waveguide. The focusing can be done by an objective lens or a lensed fiber. Due to optical reciprocity, light can be coupled out of a waveguide in a similar fashion.

By coupling light into the waveguide at one of its ends and collecting the optical output at the other one can measure the total (in-out) insertion loss of the waveguide. This includes in- and out-coupling losses into/from the optical waveguide from/to the source and the detector and any other additional losses (typically propagation losses) present inside the waveguide device. A butt coupling setup (as the one of Figure 4.9) can be easily adapted to analyze the waveguide response at different polarizations and wavelengths through the choice of suitable coupling optics. However, one practical drawback is represented by the need for in-plane optical access to the waveguide end-facets for the coupling. Furthermore, the waveguide facets have to be extremely smooth (i.e. polished to optical grade) in order to minimize scattering losses, which implies several additional processing steps.

Figure 4.9 shows the butt-coupling setup I have used for the optical characterizations of my LNOI waveguides. The output of a tunable laser, set to operate at wavelengths around 1550 nm, was coupled by a lensed fiber to the fabricated LNOI chips, while the output of the waveguides was collected by an infinity-corrected 40× optical lens and then measured by a detector. The methods were suitable for waveguide lengths of ~10 mm, whose end-faces were cleaved and then fine-polished by FIB milling.
4.2. CHARACTERIZATION METHODS

Grating coupling

The employment of butt-coupling methods has nevertheless some practical limitations, especially when addressing high device packing densities on an integrated optical chip. In these cases, for maximum compactness and flexibility in addressing multiple optical waveguides on chip, one can employ grating structures made at the ends of the waveguide to assist the coupling with optical fibers placed on top of the waveguide chip (almost vertically).

The gratings used for in-/out-coupling of the optical signal, though, need to be specifically designed and fabricated to match each waveguide, desired spectral bandwidth of operation, and polarization. The period of the grating coupler structure, \( \Lambda_c \), required for the optimal coupling of a given wavelength, \( \lambda_0 \), is given by a longitudinal phase-matching condition between the guided mode (of effective index \( N_{\text{eff}} \)) and the out-coupled radiation (see section 3.2.1), which can be expressed as [131]:

\[
\Lambda_c = \frac{\lambda_0}{N_{\text{eff}} - n_{cl} \sin \theta}
\]

where \( n_{cl} \) is the refractive index of cladding layer and \( \theta \) is the coupling angle. Deviations from the phase-matching condition of eq. 4.1, which may be induced by defects and nonuniformities in the fabrication process (such as changes of the grating period, or waveguide non-homogeneities affecting \( N_{\text{eff}} \)) can significantly degrade the performance of a grating coupler.

Figure 4.10(a) shows a setup using a grating coupler to couple optical signals into/from optical chips from/to optical fibers. The microscope objective and lensed fiber of the butt-coupling setup of Figure 4.9 are here replaced by optical fibers, aligned by micromanipulation stages on top of the waveguide gratings. A polarization controller is added to control the polarization of the input beam to match the one the grating couplers are designed for. As illustrated in Figure 4.10(b), the grating couplers are wider than the optical waveguides at the core of the device (<1 \( \mu \)m). Their widths are chosen to be 12 \( \mu \)m, to maximize the overlap with the optical mode size of single-mode optical fibers at telecom wavelengths. This requires additional integrated optical tapers on chip, to adiabatically reduce the waveguide width to the desired one in the central waveguide. Based on reciprocity, the same
setup can be used to couple out the optical signal from a similar grating coupler.

![Diagram](image)

**Figure 4.10.** (a) Optical setup used for transmission measurements using grating couplers. (b) Sketch of the planar waveguide geometries on chip, including in/out couplers, tapers and waveguide Bragg gratings. (c) Sketch of the configuration used for out-of-plane coupling from a standard telecom optical fiber into the LNOI waveguide via a grating coupler. (d) SEM image of a LNOI grating coupler.

**Loss measurement**

A key figure of merit for waveguide devices, which impacts directly their efficiency and hence power consumption in different applications, is their losses. Apart from the losses associate to input and output coupling, an optical mode travelling through a waveguide experiences propagation loss which scales exponentially with the waveguide length $L$. Due to propagation losses, the power of the signal travelling inside the waveguide is attenuated as $P_{out} = P_{in} e^{-\alpha L}$, where $\alpha$ is the propagation loss factor of the optical mode under consideration.

In my work I have employed two complementary methods to accurately determine the propagation losses in optical waveguides, in butt-coupling and grating-coupling configurations, namely: the Fabry Perot method and the cut method. They are briefly described in what follows.

**Fabry-Perot method**

The interfaces between an optical waveguide and the surrounding medium (usually air), lead to some reflection, which in turn creates a weak Fabry-Perot cavity, between the polished end-facets of a waveguide used for butt-coupling. As a result, the waveguide throughput is modulated by interference fringes and changes periodically with the input wavelength. The visibility of the
4.2. CHARACTERIZATION METHODS

Fabry-Perot fringes is directly related to the losses experienced by the optical field inside the cavity (i.e. in the waveguide). In a single-mode waveguide, the propagation loss factor can be determined through the following formula [30]:

\[
\alpha (\text{dB/cm}) = -4.34 \frac{L}{L (\text{cm})} \ln \left( \frac{1 - \sqrt{r}}{\sqrt{r} + 1} \right)
\]

where \( L \) is length of the waveguide, \( r \) is the measured ratio between maxima and minima of the output power modulation \( (r = P_{\text{max}}/P_{\text{min}}) \), and \( R \) is the reflection coefficient of the waveguide facets. The spacing between two adjacent peaks in the transmission spectrum is given by \( \Delta \lambda_{\text{FSR}} = \frac{\lambda_0^2}{2N_{\text{eff}}L} \), where \( N_{\text{eff}} \) is the effective index of the guided mode at the wavelength \( \lambda_0 \). As an example, Figure 4.11(a) presents transmission spectrum of a \( \sim 8 \) mm-long LNOI waveguide with a cross section of \( 0.3 \times 2.9 \) \( \mu \text{m}^2 \), fabricated by chemical dry etching. The measurement was performed using the butt-coupling setup of Figure 4.9.

![Figure 4.11](image)

**Figure 4.11.** (a) Fabry-Perot measurement (waveguide throughput as a function of wavelength) for a 9 mm-long, 2.9 \( \mu \text{m} \)-wide and 300 nm-thick LNOI waveguide. (b) Logarithmic plots of the transmission measured for different waveguide lengths (markers) and linear fits (dashed lines) used to extrapolate their propagation losses by the cut-back method. Data for 300 nm-thick LNOI waveguides with widths of 0.66 \( \mu \text{m} \) and 2.35 \( \mu \text{m} \) are plotted in orange and green, respectively.

**Cut-back method**

Given the exponential law of attenuation of the guided optical power with the waveguide length, propagation losses can also be quantified by measuring the transmission from waveguides with different lengths and fitting the experimental data to an exponential curve. Figure 4.11(b) illustrates examples of cut-back measurements made on waveguides with different widths (0.66 \( \mu \text{m} \) and 2.35 \( \mu \text{m} \)) and fabrication conditions. The slopes of the linear fits made on the experimental data (markers), in logarithmic scale, yield directly the propagation loss coefficients of the waveguides to be 76 dB/cm and
15 dB/cm for 0.66 µm- and 2.35 µm-wide waveguides, respectively. In section 5.4 the critical impact of waveguide design (width) and process conditions (fabrication process and adding a PMMA cladding) on the loss figure of merit will be explained.

4.2.4 Polarimetric SHG measurements

Butt-coupling in a vertical transmission microscope configuration, schematically shown in Figure 4.12, was employed for the optical characterization of nanopillar waveguides. Here, due to the reduced size (ultrahigh confinement) of the optical modes, 50× objectives were used on both sides. The nanopillars were pumped with ultrashort pulses from a Ti:sapphire laser in order to excite and investigate the polarimetric properties of their nonlinear response. Specifically, for the purposes of Paper A, I systematically investigated the polarimetric properties of the SH signal generated in individual nanopillars, as shown in Figure 4.12(a).

![Diagram of experimental setup](image)

**Figure 4.12.** (a) Sketch of the experimental setup used for polarimetric SHG measurements, with indication of the NP orientation with respect to the LN crystal-axes of LN (X, Y, Z). Example of the polarimetric response retrieved by: (b) simulations of nonlinear interactions in the nanopillars (own MATLAB code) and (c) experiments, here shown for the case of X-polarized SH in bulk Z-cut LN, with a varying in-plane (X-Y) polarization of the FF pump, $\theta$ being the FF polarization angle with respect to the X axis of the crystal.

A half-wave plate acting on the FF input allowed to control the polarization of the incident light in the (X, Y) plane (transverse plane of the nanopillar). The FF light was focused and coupled into the nanopillar through a 50×
objective lens. Another 50× objective lens was then employed to collect the output (SH and FF) light from the nanopillar, which then passed through a high-pass filter to remove the FF component. A polarizer after the high-pass filter provided also the possibility to analyze the different polarization components of the generated SH signal.

In experiments of Paper A, I followed a systematic procedure for alignment and calibration of the samples in the system. The input FF signal was first focused on an unetched area of the bulk sample, at the surface of the sample. Then polarimetric measurements (in this case with rotating incident FF polarization and a fixed SH polarization) were performed to obtain a reference response from bulk LN. The experimental settings were further adjusted and the accuracy of their results was checked against the theoretical predictions for the bulk LN crystal, shown in Figure 4.12(b), to ensure a correct focusing and relative alignment of the microscope and the LN sample (see Figure 4.12(c)). After this checks and optimizations, the sample was moved in the (X, Y) plane to couple the incoming FF beam was coupled into desired nanopillars and study their SHG response as a function of the input FF polarization.
Chapter 5

Summary of main results

This chapter provides an overview of the aspects dealt with in the publications attached to this thesis, on novel LN-based integrated optical devices (Papers A-C) afforded by the developments of nanotechnology in this material, as well as further insights in novel emerging applications of LN, where our technology platform can find further exciting perspectives. This is exemplified by Papers D-E, which employed the LN and LNOI technologies developed during this thesis to engineer templates used for novel biomolecular and biomedical applications pursued in collaborations with scientists at the Conway Institute and University of Dublin in Ireland. The two examples deal with the enhancement of Raman sensitivity for biomolecular spectroscopy (Paper D) and a controlled cell growth in microfluidic environments (Paper E).

In this chapter I also review my further progress in addressing critical issues for LNOI devices beyond the scope of the above-listed publications. This includes a systematic experimental work aiming at reducing the propagation losses of single mode LNOI waveguide at 1550 nm (~0.4 µm² cross-sections), and further technological steps for electrode-functionalization of my devices, in view of implementing enable electrically tunable devices on the in-house LN PIC platform we now have at KTH (one of the very few currently available at international level).

5.1 Paper A: SHG in LN nanopillars

The tight confinement of optical modes in submicrometric waveguides results in non-negligible longitudinal components in their guided modes (see section 3.1.1). The existence of such fields reveals itself not only in atom-photon interactions, tweezing [152] and particle acceleration [153] atop photonic wires and fibers, but also may have implications in nonlinear interactions
occurring in silicon photonic nanowires [154-156], nano-tapered optical fibers [157-159] and nanopillars in III-V semiconductors [106, 160-162]. Paper A deals with these questions in the context of LN and its second-order nonlinearity in waveguide configurations. The guiding structures here are made of micrometer-high LN nanopillars with diameters as low as 150 nm carved into LN crystals by refined FIB milling. The high resolution and quality of the NP structuring allowed to sample and fully characterize the impact of the longitudinal guide-wave field components on the polarimetric response of LN devices. By backing up systematic fabrication and optical characterization efforts in the lab with a coupled-wave multimode analysis of SHG in the nanopillars (with ad-hoc models implemented in COMSOL and Matlab codes), I could obtain excellent agreement between theory and experiments. Furthermore, I experimentally demonstrated the possibility to substantially reshape the SHG polarimetric response of LN, by using the FIB fabrication capabilities developed in this thesis, opening up further perspectives for metamaterial studies and advanced nanoscopic probes for imaging at LN surfaces (discussed in the conclusions).

Details of the work are provided in Paper A; here the general principle of operation of the NP SHG devices is briefly illustrated. Technical details on the modal analysis of SHG in LN nanopillars are provided in Appendix B. A sketch of the physics underlying the size-tunable polar response of the SHG nanopillars is illustrated by Figure 5.1.

The tight confinement in the NPs (with diameters less than 350 nm) results in the presence of guided modes with an appreciable longitudinal component of the electric field (\(E_z\), along the NP axis), in addition to the “conventional” transversely polarized component. By using suitable \(\chi^{(2)}\) tensor components (i.e. suitably orienting the NP axis with respect to the crystallographic axis), the presence of \(E_z\) (say at the FF) can be revealed through nonlinear studies. This results in the generation of SH polarization components “not allowed” by SHG in the bulk, i.e. with conventional TEM free-space beam excitation. NPs of varying diameters (from 150 to 325 nm) in different LN cuts (\(X, Y, Z\)) and also on LNOI substrates were made by the refined FIB milling technology described in section 4.1.1. By using the experimental setup and methodology presented in section 4.2.4, their response was systematically characterized for varying polarizations of the FF (and also of the SH). Theoretical modelling based on the equations and methodology outlined in Appendix B was used to perform and compute the modal structure of the NP at the FF and SH. Vectorial modelling in COMSOL Multiphysics® allowed to determine the field distribution for all components of the electric field \((E_x, E_y, E_z)\) and the effective indices for each mode at the FF and SH. A coupled mode analysis was then developed to analyze the SHG response in the guided-wave configuration, isolating the contributions of each mode and its polarizations. Finally, the experimental results (SH polar plots)
were compared to the theory finding very good agreement between theoretical predictions and experiments.

![Figure 5.1](image-url)

**Figure 5.1.** Tilted (52°) SEM image of a 325 nm NP in Z-cut LiNbO$_3$. Examples of calculated transverse ($x$, $y$) distributions of the transverse ($E_T$) and longitudinal ($E_Z$) field components supported by a 275 nm NP; (b) $HE_{11}^{(x)}$ mode at $\lambda_{FF} = 850$ nm; (c) $HE_{11}^{(x)}$, $TE_{01}$, $HE_{21}^{(x)}$, and $TM_{01}$ modes at $\lambda_{SH} = 425$ nm. (d) Theoretical polar plots of $X$-polarized SH intensity in a Z-cut NP as a function of the input FF polarization angle $\theta$ (see the inset), calculated for the two limit cases of purely transverse (red) and longitudinal-transverse (blue) SHG coupling. Experimental Polar plots for $X$-polarized SHG in Z-cut NPs with diameters of (e) 275 nm and (g) 175 nm show how polarimetric response of each NP can change. Solid lines: numerical fits based on Eq. (2) in Paper A.

## 5.2 Paper B: birefringence-free LN waveguides

As briefly mentioned in section 3.1.3, birefringence can be an undesired side-effect arising even in isotropic materials as a consequence of asymmetric waveguide cross sections [134, 140]. Several methods have been considered to
address this issue employing approaches adding further fabrication complexity, such as adding geometrical anisotropies by etching or layered growth and inducing strain [132, 163] to compensate the waveguide anisotropy.

LN is by itself a birefringent material. In Paper B, we show how this feature can be effectively exploited to counterbalance waveguide modal birefringence, relaxing the tight fabrication constraints required to achieve polarization-independent effective indices for the fundamental modes (TE\textsubscript{00} and TM\textsubscript{00}) in high confinement waveguides. The principle is illustrated by Figure 5.2.

The waveguide birefringence is controlled by the ration between the waveguide width (w) and height (h). Assuming for instance a uniform and isotropic cladding and typical LNOI thicknesses for single mode operation in the near-infrared and telecom range (\(\lambda \sim 1550\) nm), in order to ease fabrication, one would like to achieve birefringence free operation with \(w > h\). In this regime, in the absence of material birefringence, the effective index of the TE modes is higher than the one of the TM ones: \(N_{TE00} > N_{TM00}\). The negative uniaxial birefringence of LN (\(n_e < n_o\)) can then be effectively used to counteract the waveguide birefringence by making a waveguide where the TE\textsubscript{00} mode is aligned to the Z crystal axis of LN. Extensive simulation analyses (see Paper B) conducted for both ridge and buried waveguides, indicating the possibility to use this effect and achieve broadband polarization-independent effective indices for the fundamental modes in LNOI waveguides with realistic design parameters (e.g. \(w \sim 400\) nm, fully feasible with our LNOI technology).

Furthermore, we found working points in the LNOI design where polarization-independent phase and group velocity could be achieved with fundamental modes, as highlighted by the diagrams in Figure 5.3. This peculiar feature, interesting to make polarization-insensitive devices
operating also in the pulsed regime, has not, to the best of our knowledge, been highlighted in any other nanophotonic material platform to date. [133].

Figure 5.3. Regions in the two-dimensional parameter space defined by wavelength and waveguide width, where the difference of the $TE_{00}$ and $TM_{00}$ mode effective (blue) and group (red) indices is below 0.001 for: (a) ridge and (b) buried waveguides in 300 nm-thick Y-cut LNOI. Plots reprinted from [133].

5.3 Paper C: waveguide Bragg gratings in LNOI

Bragg gratings are essential elements in integrated optical circuits, thanks to their applications as optical filters, dichroic reflectors and distributed sensing elements. Waveguide Bragg gratings have been widely investigated in different optical platforms such as optical fibers [164, 165], silicon [12, 107] and other semiconductors [166]. They can potentially benefit from even more advanced functionalities when realized on a material such as LN, where electro- and nonlinear-optical effects can make them tunable with applied voltages or optical fields.

Prior to the advent of LNOI platform, several attempts were performed to realize Bragg grating structures in relatively large-mode (weakly guiding) LN waveguides [167-171], by different methods such as thermally fixed photorefractive gratings, laser writing, FIB milling or EBL patterning. In most of these attempts, the investigated Bragg structures yielded rather low extinction ratios, i.e. the contrast between the grating transmission inside and outside the bandgap. This was mostly due to the low index contrast of the employed waveguides. Moreover, in all cases, extra fabrication steps needed to be performed to realize Bragg structures separately after waveguide fabrication. Table 5.1 summarizes extinction ratios obtained in these early approaches by other groups, together to the results achieved in this thesis (Paper C and further improvements on that described in the next section, unpublished).
Table 5.1. Examples of extinction ratios achieved in waveguide Bragg gratings fabricated over LN platform.

<table>
<thead>
<tr>
<th>Platform</th>
<th>Optical mode</th>
<th>Extinction ratio</th>
<th>Bragg fabrication</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti-indiffused LN</td>
<td>$TE_{00}$</td>
<td>11 dB</td>
<td>Thermally fixed photorefractive gratings</td>
<td>[167]</td>
</tr>
<tr>
<td>Ti-indiffused LN</td>
<td>$TM_{00}$</td>
<td>7 dB</td>
<td>Proton-exchange to induce gratings</td>
<td>[168]</td>
</tr>
<tr>
<td>Proton exchanged LN</td>
<td>$TE_{00}$</td>
<td>5.7 dB</td>
<td>EBL and etching on predefined waveguides</td>
<td>[172]</td>
</tr>
<tr>
<td>Ti-indiffused LN</td>
<td>$TM_{00}$</td>
<td>20 dB</td>
<td>FIB milling on predefined waveguides</td>
<td>[173]</td>
</tr>
<tr>
<td>Diced Ti-indiffused LN</td>
<td>$TM_{00}$</td>
<td>18 dB</td>
<td>FIB milling on predefined waveguides</td>
<td>[174]</td>
</tr>
<tr>
<td>LNOI</td>
<td>$TE_{00}$</td>
<td>27 dB</td>
<td>Gratings and waveguides fabricated simultaneously</td>
<td>Paper C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LNOI</td>
<td>$TE_{00}$</td>
<td>45 dB</td>
<td>Gratings and waveguides fabricated simultaneously, measurement setup modified</td>
<td>Here</td>
</tr>
<tr>
<td>LNOI</td>
<td>$TE_{00}$</td>
<td>14 dB</td>
<td>Gratings and waveguides fabricated simultaneously</td>
<td>[82]</td>
</tr>
</tbody>
</table>

The goal I set for myself in this thesis was to develop integrated high-extinction Bragg grating devices in LNOI, with small footprints and sub-micrometric structures, including not only Bragg reflectors, but also fiber grating couplers, tapers and waveguides all fabricated simultaneously, during a single EBL patterning process. Despite the challenge of optimizing the etching processes for such a complex architecture, I proved this to be possible, achieving record Bragg extinction ratios in transmission, of up to 27 dB (limited by the measurement sensitivity), as reported in Paper C. This approach is also –among the ones listed in Table 5.1– the only one truly viable for process upscaling to larger chips and even wafer sizes, similarly to what achieved on Si-photonic foundries.

5.3.1 Fabricated LNOI structures

The waveguides used for Paper C were made following the fabrication steps described in section 4.1.2, on commercially available (Partow Inc.) Y-cut LNOI bonded to a silicon carrier wafer. A main drawback of such substrates (some
of the first commercially available at the time) was the relatively poor quality of the LNOI wafer, exhibiting quite high propagation losses already without any processing (3 dB/cm, measured by prism coupling). Subsequent structural and wet etching studies revealed also the presence of quite high strain in the thin-film LN layers.

The originally 300 nm-thick LNOI substrate was dry etched to a depth of ~270 nm to realize multiple integrated optical devices with the architecture shown in Figure 4.10(b) on a single chip. All waveguides were aligned along the X crystallographic axis of the LN layer. The optical signals were coupled into and out of the LN optical chip through fiber grating couplers with periods of \( \Lambda_C = 1.24 \, \mu \text{m} \) (see section 4.2.3). The waveguide grating couplers were designed and optimized with simulations performed with a Lumerical® software.

Even if the main goal for the research presented in Paper C was to realize waveguide Bragg gratings with high extinction ratios, the performance of the grating couplers is worth of notice. Indeed, there are only few reports of grating couplers in LNOI platform, with additionally relatively low coupling efficiencies. The grating couplers described in Paper C do not employ any reflective layer (in contrast to e.g. [73, 74]), and yet can couple optical signals with wavelengths of ~1550 nm into the \( TE_{00} \) guided mode of submicrometric waveguides with a total insertion loss below 10 dB. Furthermore, the 3 dB-bandwidth (i.e., the bandwidth after which the insertion loss degrades by 3 dB) of these grating couplers is also wider than 80 nm (essentially limited by the tunability range of our laser rather than the grating itself).

The waveguides of Paper C were made by dry etching and had sidewall angles of 45°. The resolutions reached in the fabrication process allowed me to reliably make fine-pitch gratings in them (~500 nm period and feature sizes down to 100 nm) and even investigate different profiles of the sidewall modulation used to make the \( TE_{00} \)-mode Bragg gratings. This is illustrated by the SEM images of Figure 5.4, for (a) rectangular and (b) sinusoidal width corrugations. For The purpose of mapping out the full parameter space of the Bragg grating response, multiple devices were simultaneously made on a single chip, with different corrugation depths (\( \Delta w = 50, 75, 100, 125 \, \text{nm} \)) over an average waveguide width of 760 nm.

Bragg gratings of lengths up to 0.5 mm were fabricated with essentially no stitching errors. Nevertheless, occasionally lateral misalignments, such as the one shown in Figure 5.4(b) (leading to a non-perfect centering of the Bragg grating with respect to the waveguide) occurred in the EBL writing, due to a shift of the EBL Raith system when switching from the FBMS-writing modality used for the straight waveguides and to the area-writing modality used to achieve a rectangular modulation of the grating. However, in accordance with the predictions of the simulations made posteriori for such structures using Lumerical®, this shift did not degrade the performance of the Bragg gratings in terms of the extinction ratios achieved in the experiments.
In parallel, the cut-back method described in section 4.2.3 was employed to evaluate the propagation losses inside the waveguides with additional test structures. For this purpose, similar waveguides with widths of $w_0 = 760$ nm and different lengths were fabricated. The propagation losses of the fabricated waveguides were found to be quite high (76 dB/cm). This was partially due to the residual roughness associated with the etching recipes used at the time, further improved since then with a new optimized process (see section 4.1.2). Moreover, the effect of losses is here exacerbated by the relatively small cross-sections ($\sim 0.3 \times 0.76 \, \mu m^2$) and almost complete etching of the LN layer (ridge waveguide geometry). These effects can be alleviated by resorting to rib waveguides and by increasing the waveguide widths, although at the expenses of a less tight mode confinement in the former case and of multimodality of the waveguide in the latter case. Another viable and effective approach to reduce the propagation loss in the fabricated waveguides, for which we have new results (see also section 5.4) is the employment of a cladding layer, such as PMMA or SiO$_2$, in analogy to loss-reduction strategies used for Si-photonics devices.

Last but not least, the intrinsic propagation losses of the as-purchased slab LNOI films significantly contributed to the high propagation loss of the devices made for Paper C and we could subsequently significantly improve on that by buying new and better wafers which became available at a later stage. Another possibly important factor contributing to the rather high propagation losses observed in the experiments of Paper C was the use of a Si carrier wafer for the LNOI. The large mismatches between Si and LN (in terms of their crystal structures and thermal expansion coefficients) can be expected to further increase the propagation loss of the LNOI device during its subsequent processing, given the multiple temperature cycles that the samples have to go through during waveguide fabrication. These effects can be eliminated by using LNOI on LN instead of Si carrier wafers.
5.3.2 Bragg grating performance

In Paper C, the performances of the Bragg gratings were simulated based on Transfer Matrix Method (TMM), described in Appendix A. In our approach, transverse profiles of the guided modes in different sections of the Bragg grating were first simulated using COMSOL Multiphysics® software. I then developed a MATLAB code based on TMM formulations in order to theoretically calculate performance characteristics of the fabricated Bragg gratings, matching well the experimental results.

As discussed in Paper C, the presence of a propagation loss of 76 dB/cm significantly reduces the extinction ratios obtainable in a Bragg grating. This inclusion of losses in the model confirmed this effect. However, a discrepancy was still observed between the measurements and the simulations. As speculated in Paper C, this discrepancy may be due to additional scattering by grating imperfections (not accounted for in the model I used) or to the limited detection sensitivity of the experimental setup. Indeed, after Paper C, we built a new measurements setup and refined further the measurement software and procedure, significantly improving the detection sensitivities. As shown in Figure 5.5, by repeating the measurements on the devices of Paper C with this new setup we could indeed reach in the experiments maximum extinction ratios of 45 dB matching well the values predicted by simulations. This is a further proof of the excellent quality of the fabricated Bragg gratings.

Figure 5.5. Normalized transmission spectra of rectangular Bragg gratings measured of different values of the width corrugation, $\Delta \omega$ (50, 75, 100 and 125 nm, color-coded in the legend). (a) detection-limited data published in Paper C (experiments in 2017) and (b) experimental results obtained from the same sample after improving the measurement sensitivity, showing contrast ratios up to xx in the Bragg grating response.

5.4 Reducing propagation losses in LN waveguides

Reducing propagation losses in waveguiding structures is a key challenge to overcome, in order to develop functional integrated optical platforms. Losses weaken the optical signals, and significantly affect nonlinear, electro-optical
and filtering performances of such devices. As previously mentioned in section 04.1.2, the etching roughness and the associated propagation losses in LN waveguides can be significantly reduced by resorting to Ar-ion milling instead of fluorine-based dry etching processes. In this section, I will provide some not published results achieved with additional strategies and further steps taken to reduce the propagation losses in our LNOI waveguides.

It is worth mentioning at this point the record low value, 0.027 dB/cm, recently reported by M. Lončar’s group at Harvard for the propagation losses of LNOI waveguides (also made by Argon-ion milling), at telecom wavelengths [65]. These are impressive results in the context of LN. Nevertheless, they were obtained in relatively large waveguides (widths of 2.4 µm and partially etched rib waveguide in 600 nm-thick LNOI), which are highly multimode. This still leaves the reduction of propagation losses in single-mode LNOI structures as an outstanding challenge.

5.4.1 Progress in reducing propagation losses during this thesis

The results of our first work on LNOI waveguides etched with fluorine-based plasma, described in section 04.1.2, were reported in conference contribution B. The propagation losses in these devices (measured by the Fabry Perot method) amounted to 8.62 dB/cm for ~2.9 µm-wide, 260 nm-thick LNOI waveguides (Partow Inc. wafers, as in Paper C), supporting several optical modes. The same dry etching technique, combined with EBL, enabled us to realize submicrometric single-mode LNOI waveguides. However, in this case the propagation losses (with the same LNOI substrate) increased to ~80 dB/cm for 800 nm-wide waveguides, as shown in Figure 4.6(d).

To improve on these results, I have resorted to Argon milling instead. The first achievements (waveguides used in Paper C), however, did not seem very promising in terms of loss reduction, yielding similar values, namely 76 dB/cm for 760 nm-wide LNOI waveguides.

With further optimizations of the Ar-ion milling process, as discussed in section 4.1.2, I could realize integrated optical LNOI devices with much smoother sidewalls. Additionally, having recognized the problems with the original LNOI wafers, we moved to the ones of a different provider, this time using LN carrier wafers. An example of the last generation of devices is illustrated in Figure 4.6(f). This has provided promising results. At wavelengths close to 1550 nm, the propagation losses inside 0.86 µm-wide, 500 nm-thick waveguides have decreased to 10.6 dB/cm. Furthermore, the propagation losses in the same waveguides dramatically decreased to 1.13 dB/cm, when the waveguides were cladded by a PMMA layer. Figure 5.6 shows the fiber-to-fiber transmission spectrum of a waveguides with lengths of 500 µm, as well as data used for calculation of propagation losses in such waveguides using cut-back method.
Figure 5.6. (a) Fiber-to-fiber spectral transmission response of a 0.86 µm-wide, 500 nm-thick, 0.5 mm-long waveguide; (b) logarithmic plots of the transmission measured for waveguides similar to that of part (a), but with different lengths (markers) and linear fits (dashed line) used to extrapolate their propagation losses by the cut-back method. Values for markers and error bars are obtained by calculating average and standard deviations of transmission response of each waveguide at the spectral range of 1550±20 nm.

5.5 Paper D: photo-induced Raman enhancement in LNOI

The technology I have developed and employed in this thesis not only can be employed for fabrication of integrated optical chips and elements, but also provides us the opportunity to fabricate desired structures on LN and LNOI platforms for further interdisciplinary studies on different physical, chemical and biological phenomena. As an example, the samples provided by us have been explored by our collaborators in University College Dublin to study Photo-Induced Enhanced Raman Spectroscopy (PIERS), summarized in Paper D. An enhanced Raman spectroscopy can be used for a broad application range, from geology [124] to semiconductor surface analysis [125] to life sciences [126].

The extremely low efficiency of Raman scattering process can be enhanced by using metallic nanostructures –employed in Surface-Enhanced Raman Spectroscopy (SERS)– which may be deposited by means of ferroelectric lithography on patterned bulk LN templates [94]. In the work of Paper D, we expand further on these results, exploring the potential of LNOI substrates for enhancing Raman sensitivities by physical and chemical methods taking further advantage of photonics. In particular, Paper D shows how the combination of field enhancement due to dielectric effects (at the multiple interfaces of the LNOI structure), in combination with the enhancement of scattering and plasmonic effects at the surface of –deliberately roughened– LNOI surfaces fabricated by dry etching, can result in significant enhancements in Raman sensitivities on biological specimen.

The paper also shows how Raman signals can be further enhanced in PIERS, where the sample is pre-irradiated by a UV source. As a consequence of UV pre-irradiation, free carriers are photogenerated in LN and can be
transferred to metallic nanoparticles (in contact with the analyte) present at the LN surface, resulting in further enhancement of the Raman scattering signal. Figure 5.7(a) schematically shows the PIERS process on a LN substrate with pre-deposited silver (Ag) nanoparticles. Upon pre-irradiation of the sample by UV light with energy higher than the energy bandgap of LN, free electrons are generated on the LN surface and then transferred to the Ag nanoparticles which in turn results in further enhancement of Raman signals.

Figure 5.7. (a) PIERS in LNOI platform: pre-irradiation of the LN patterned sample creates photogenerated carriers at the LN surface which can be transferred to Ag nanoparticles and enhance the Raman signals. (b) Roughness of the etched LN surface may further enhance the scattered signal. (c) Dielectric interfaces in an LNOI sample can be employed to enhance Raman response at specific wavelengths.

The results summarized in Paper D demonstrate not only the suitability of LN as a substrate for PIERS phenomenon, but also the additional enhancement effects which are available by refractive index engineering in LNOI substrates. Specifically, the interference effects associated with the multiple reflections from the high index contrast LN/SiO$_2$ and SiO$_2$/Si interfaces in an LNOI substrate (see Figure 5.7(c)) can be used to maximize the Raman response at specific wavelengths.

Although not desirable in many photonic applications, the roughness of etched LN surfaces can be helpful in PIERS experiments. In this case, the increased scattering caused by rough etched LN surface may also prevent the organic molecule from photodegradation, by limiting the produced heat. Figure 5.8(b) demonstrates the intensity of Raman signals at a specific band (with a wavenumber of 1583 cm$^{-1}$) for in-situ Raman experiments on Ag-deposited unetched 300 nm-thick Y-cut LNOI and etched 130 nm-thick X-cut LNOI samples.

The results presented in Paper D not only prove for the first time the suitability of LNOI for PIERS and in general Raman sensing, but also pave the way for future devices employing tailored multilayer LNOI templates and potentially also LNOI waveguides for enhanced Raman spectroscopy with integrated optical structures, similar to that presented in [175, 176].
5.6 Paper E: control of neuron growth with patterned LN templates

The interest at new biocompatible and functional materials for biological applications is driven by the need to understand how materials with different surface properties may influence and/or control the cellular response. This can be helpful in order to design tissue-engineered constructs, biological platforms or implantable devices. Surface features in general affect cell growth and proliferation. It has been previously shown that axons tend to align and accelerate along longitudinal reliefs with specific width and depth combinations on different substrates [177, 178]. Moreover, there have been studies showing that electric fields can also affect their growth direction [179].

Meanwhile, it has already been shown that uncoated LN is biocompatible [99] and can be a suitable candidate for cell growth studies, providing an intrinsic electric field on its surface which can further be combined with topographic structures on its surface [180]. Furthermore, other characteristics of LN such as photovoltaic, pyroelectric and acoustic properties can also be exploited for patterning and sorting biological cells [98].

In Paper E we have, in collaboration with researchers in University College Dublin, integrated the LN structuring technology (presented in this thesis) into microfluidic devices to provide topological cues for axons isolated from their cell bodies. For these investigations, the fabrication technology described in section 4.1.2 was employed to realize 550 nm-deep structures on bulk LN. The study was complemented by investigating also the effect of

Figure 5.8. (a) Raman spectra of an organic molecule on X-cut LNOI (green) and X-cut LN (black) showing the enhancement associated to the optical multilayered structure of the LNOI template; (b) background-subtracted intensity for the 1583 cm⁻¹ band for in situ radiation for the organic molecule on Ag-deposited etched X-cut LNOI (black) and Ag-deposited unetched Y-cut LNOI (red). Reprinted with permission from R. Al-Shammari et al., “Photo-induced enhanced Raman from lithium niobate on insulator template”, ACS Applied Materials & Interfaces 10(36), 30871-30878 (2018). Copyright 2018 American Chemical Society.
surface charges, with the growth of axons on a PPLN sample. Although the investigations reported in Paper E are performed on bulk LN crystals and no specific optical functionality was associated to the topographic structures, a potential further extension consists of adding ridge waveguides to the sides of axon-guiding grooves and develop waveguide-based devices for biological sensing of cells and monitoring their growth \cite{181,182}.

Figure 5.9 depicts some of the results summarized in Paper E. As it can be seen, the axons typically elongate parallel to micro-grooves at the LN surface (Figure 5.9(a)) and preferentially align by the edges. Further results in Paper E demonstrate that, if isolated from their cell bodies through microfluidic chambers, the axons tend to navigate through two-dimensional pit arrays in a similar fashion (Figure 5.9(b)).

![Figure 5.9](two_images.png)

**Figure 5.9.** Alignment of neuronal axons (fluorescently labelled in the confocal images) with topographic features of selectively etched LN substrates in: (a) 9 mm-long micro-grooves (horizontal lines in the picture), and (b)-(c) hexagonal pits (whose edges are marked in green). Images reprinted from **Paper E**. Reprinted with permission from R. D. Kilinc et al., “Charge and topography patterned lithium niobate provides physical cues to fluidically isolated cortical axons", Applied Physics Letters **110**(5), 053702 (2017). Copyright 2017 American Institute of Physics.

The results provided in Paper E suggest LN as a promising substrate for neural studies. Patterned LN may be used as a template for complex cell culture lab-on-a-chip devices and combined with the nanoscopic excitation and sensing potentially affordable with the LN photonic integrated circuits technology developed in this thesis.
Chapter 6

Conclusions and future work

In this chapter, I will provide a conclusive overview of results summarized in this thesis, followed by a review of some possible pathways for further extension of the work summarized here.

6.1 Conclusions

In this thesis, I have developed and optimized fabrication technologies, design and analysis tools, for photonic devices with applications on the LN platform, namely polarization control in blue second harmonic emission, waveguide Bragg-grating rejection filters and integrated fiber-couplers operating at telecom wavelengths, and broadband polarization-insensitive waveguides. Furthermore, I have employed some of the outcomes of the fabrication pipeline to realize LN-based devices for other, interdisciplinary, applications, namely Raman sensing and controlled cell growth.

6.1.1 Integrated optics in LN

By providing excellent intrinsic electro- and nonlinear- optical properties in a wide transparency spectrum, LN remains a material of great interest for the realization of ultrafast optical devices such as electro-optical modulators and frequency converters.

Advances in nanofabrication technology are now rapidly driving the evolution of LN devices into sub-micrometer sized ones, taking advantage of the high index contrast and maturity now offered by commercially available LNOI wafers. The tight confinement of optical modes in such sub-micrometric devices, in turn potentially yields a number of very attractive features, associated with strong longitudinal components which may affect (control) the polarization response of nonlinear interactions, enhance the coupling
strengths in waveguide grating devices, provide new possibility for dispersion and birefringence engineering. Moreover, the small cross section areas of the guided modes in such devices can lead to enhanced electro- and/or nonlinear-optical responses inside them.

With this thesis I built up a toolbox which brings us a few steps closer to the ultimate goal of realizing fully-functional integrated optical circuits in LN, similar to the nanophotonic capabilities available in Si-photonics and demonstrated along the way a number of novel devices. The results summarized in Paper A show how the fine-tuning of the geometry of LN nanopillars by FIB techniques and tight confinement of guided optical modes inside them can radically change their nonlinear response compared to the bulk or to weakly-confining conventional waveguides in LN.

The theoretical results summarized in Paper B show how an appropriate choice of geometrical properties, cladding layer, and orientation with respect to the LN crystalline axes, may be used to achieve polarization-insensitive LNOI waveguides in a broad spectral range, and even in pulsed operation regimes.

The first-time realization of waveguide Bragg grating reflectors in LNOI is another achievement, reported in Paper C. The same work presents also a technology to make several integrated optical elements, including grating couplers, waveguides and Bragg gratings simultaneously through a single fabrication process on LNOI chips. The grating couplers exhibit a total coupling loss below 10 dB over a wide band, exceeding 80 nm (and limited by the tunability of our laser) at wavelengths around 1550 nm. The waveguide Bragg gratings, in turn, can provide extinction ratios of up to 45 dB, a record for LN. Moreover, theoretical and experimental investigations of such devices are in very good agreement and show how parameters such as shape, length and corrugation depths of Bragg gratings can affect their performance characteristics (extinction ratio and bandwidth).

The reduction of propagation losses inside single-mode LNOI waveguides at telecom wavelengths is another major achievement reported here. With a consistent improvement of the fabrication process employed to realize LNOI waveguides, we can now make low-loss single mode waveguides with propagation losses of only 1.13 dB/cm. This low propagation loss is critical in reducing the power consumption of LN-based PIC elements and allows us now to realistically envisage to add electro-optic and nonlinear optical functionalities to them, as briefly discussed in section 6.2.

### 6.1.2 Life science applications of LN

In Paper D we have shown the important role of the high index contrast provided by LNOI templates in the enhancement of Raman signals, an effect not offered by bulk LN. Furthermore, in contrast to the optical applications discussed in section 6.1.1, the deliberate introduction of surface roughness by etching proved instrumental to prevent photodegradation of the employed
organic materials, further enhancing Raman sensitivities of the nanopatterned LNOI templates.

Surface topographical structures fabricated in LN by etching have also been employed in Paper E in order to control proliferation and growth direction of axons. The effect of surface charges offered by periodically poled samples has also been studied for the same aim. The results show how selective etching and ferroelectric domains at LN surfaces integrated with microfluidics can control cell growth, without the complications implied by some other methods, involving e.g. the application of external electric fields to direct cell growth path.

6.2 Future work and perspectives

In parallel with the work of this thesis, in the last 2-3 years, a number of PIC devices have been demonstrated on LNOI, including ring and racetrack resonators [64-66], micro-disks [67-69], ultrafast modulators and switches [76-78], wavelength converters [61, 69, 79-81], and tunable filters [82]. This proves the undeniable interest and potential of this technology, which can be expected to further grow in the next few years, with new demonstration of multifunctional nanophotonic components based on LN.

A natural direction following up on the work on nanopillar SHG sources of Paper A is to use them as localized nanoprobes for bioimaging, but also – by reversing the nonlinear interaction – as parametric sources for entangled photon pairs for quantum optics applications. These studies can be further expanded to investigate nonlinear interactions in arrays of nanopillars (similar to the fabricated ones shown in Figure 6.1) and employ them to shape SH beams or act as localized nano-antennae.

![Figure 6.1](image_url) SEM images of 2D arrays of 1 µm-long nanopillars in LN, with a period of 850 nm and NP diameters of (a) 350 nm and (b) 250 nm, fabricated by FIB milling. 1 µm scale bar.

As for the planar integrated devices in LNOI, I have developed all the further fabrication steps (discussed in section 04.1.2) to realize electro-optically tunable optical devices. Shortly after our demonstration of Bragg grating devices in LNOI, electrically tunable Bragg gratings have been
demonstrated with a first proof-of-principle device [82]. Nevertheless, the performance of the latter (in terms of switching voltage of 100 V, spectral tenability of 23 pm/V and extinction ratio of 14 dB) still leaves ample room for improvement to attain the theoretical limits of the LNOI platform. To this aim, I am currently working on implementing our second-generation of Bragg grating devices with high aspect ratios and narrow bandwidths, and optimized waveguide and electrode geometries in order to reduce their required driving voltage. Electro-optical modulators, phase shifters and tunable filters realized through Bragg gratings as well as ring and racetrack resonators (similar to those shown in Figure 6.2, also fabricated during this thesis) are examples of possible architectures where the electro-optical properties of LN can also be combined with tight field confinement for switching and tuning of classical linear and nonlinear devices as well as single photon manipulation on LNOI chips.

The next step will consist of developing a periodic poling technology ad-hoc for our LNOI waveguide architecture, as join in the race on developing novel multifunctional devices on LNOI, by combining confinement in PIC structures, with electro-optic tenability and $\chi^{(2)}$ optical nonlinearities. The possibilities for novel ultrafast and compact nonlinear devices there are endless, once a combined and optimized technology becomes available. In this respect, it is worth highlighting the very recent demonstration of periodically poled LN photonic wires by the Harvard and Stanford groups [183], which achieved intrinsic efficiencies of 2600 %W$^{-1}$cm$^{-2}$, very close to the theoretical limit. This is two orders of magnitude improvement compared to traditional PPLN waveguides and paves the way for many new nonlinear devices to come.

Finally, further research directions initiated with this thesis concern the realization of hybrid devices using this time LNOI as a PIC substrate to be integrated with Superconducting Single Photon Detectors (SSPD), directly deposited and patterned on LNOI waveguides, similar to the work done on Si-nanocircuits [184, 185]. This can potentially provide unique capability for
generating and manipulating entanglement and single photon detection in a single integrated quantum optic circuit. The image on the front cover of the thesis is an example of the work currently in progress as a collaboration between our group and the one of Prof. Val Zwiller at KTH, to efficiently integrate their SSPDs on our LNOI waveguide circuits.

Last but not least, the LNOI waveguide technology and the interdisciplinary applications of LN pursued in Papers D and E provide also a number of interesting perspectives for future work. LNOI waveguides may be combined with the properties studied in Paper D and used for on-chip Raman spectroscopy, which can in turn be useful for biological applications such as real-time characterization of surface species, similar to that reported in [176]. Sensing biological particles through the evanescent fields of LNOI optical waveguides, as well as employment of optical waveguides for real-time control and monitoring of cell growth in microfluidic environments integrated with the LN optical platform are other possibilities for future devices for life science applications which may be realistically envisaged following the work of this thesis.
CHAPTER 6: CONCLUSIONS AND FUTURE WORK
Appendix A

Theory of waveguide Bragg gratings

A.1. Coupled-mode analysis

As discussed in section 3.2, if two optical modes $\mu$ and $\nu$ with propagation constants of $\beta_\mu$ ($= |\beta_\mu|$) and $\beta_\nu$ ($= |\beta_\nu|$), respectively, travel inside a waveguide Bragg grating, they can be coupled through the gratings, following the coupled mode equations [131]:

\[
\pm \frac{d}{dz}A_\mu(z) = -j\kappa^* A_\nu(z) e^{-j2\Delta z} \quad \text{A.1}
\]

\[
\pm \frac{d}{dz}A_\nu(z) = -j\kappa A_\mu(z) e^{+j2\Delta z} \quad \text{A.2}
\]

where $A_\mu$ and $A_\nu$ are the amplitudes of the modes $\mu$ and $\nu$, respectively, and phase mismatch is defined as:

\[
2\Delta = \beta_\mu - (\beta_\nu + qK) \quad \text{A.3}
\]

Considering contradirectional coupling of modes via gratings, i.e., $\beta_\mu > 0$ and $\beta_\nu < 0$, plus and minus signs should be taken for the left-hand sides of eqs. A.1 and A.2, respectively. Solving these equations with boundary conditions of $A_\mu(0) = 1$ and $A_\nu(L) = 0$ (where mode $\nu$ is the reflection of mode $\mu$, incident to a grating structure with length $L$), one can obtain the following relations:

\[
A_\mu(L) = e^{-j\Delta L} \frac{\sqrt{\kappa^2 - \Delta^2}}{\sqrt{|\kappa|^2 - \Delta^2} \cosh\{L\sqrt{|\kappa|^2 - \Delta^2}\} - j\Delta \sinh\{L\sqrt{|\kappa|^2 - \Delta^2}\}} \quad \text{A.4}
\]
A.2. Transfer Matrix Methods

Treatment of Bragg gratings with coupled-mode theory (as discussed in section A.1) is widely used to study such structures. However, this method suffers from a limitation: it assumes that existence of Bragg gratings does not change the effective refractive index of the waveguide, i.e., this method is suitable for weakly-coupled Bragg gratings (|\kappa L| < 3 [165]). Here, I will briefly describe the use of Transfer Matrix Method (TMM) for analyzing Bragg gratings. Readers are referred to [187] for a more detailed explanation. Using this approach, one can also easily introduce losses to the propagating modes, for a more detailed study of Bragg waveguide gratings.

Let’s consider the distributed feedback structure schematically shown in Figure A.1. Compared to the schematic representation of Figure 3.3, here wider (denoted here by “+”) and narrower (denoted here by “−”) parts of the Bragg grating are distinguish. The “\(\mu\)” and “\(\nu\)” indexing is also employed to distinguish between forward and backward propagating optical modes,

\[
A_\nu(0) = \frac{-j\kappa \sinh \left( L \sqrt{|\kappa|^2 - \Delta^2} \right)}{\sqrt{|\kappa|^2 - \Delta^2} \cosh \left( L \sqrt{|\kappa|^2 - \Delta^2} \right) - j\Delta \sinh \left( L \sqrt{|\kappa|^2 - \Delta^2} \right)} \quad \text{A.5}
\]

One can thus define the power transfer, via reflection, efficiency as:

\[
R = \frac{|A_\nu(0)|^2}{|A_\mu(0)|^2} = \left[ 1 + \frac{1 - \Delta^2 / \kappa^2}{\sinh^2 \left( L \sqrt{|\kappa|^2 - \Delta^2} \right)} \right]^{-1} \quad \text{A.6}
\]

Hence, one can see that maximum reflection occurs if \(\Delta = 0\):

\[
R_0 = \tanh^2 (|\kappa| L) \quad \text{A.7}
\]

The condition for maximum power reflection to occur, i.e., \(\Delta = 0\), can be obtained by substituting \(\beta_\nu = -\beta_\mu\) in eq. A.3. Thus, for first order grating to be phase-matched (\(q = 1\)), the condition can be written as \(K = 2\beta_\mu\), yielding to a grating period of \(\Lambda = \frac{\pi}{\beta_\mu} = \frac{\lambda}{2N_{\text{eff}}}\). This condition, however, occurs only for a specific wavelength (in which \(R = R_0\)). The full-width-at-half-maximum (FWHM) bandwidth of the grating coupler can thus be defined by the wavelengths in which \(R = R_0/2\), and can be derived as [12, 186]:

\[
\Delta \omega_{\text{FWHM}} = \frac{2\kappa c}{N_{\text{eff}}} \quad \text{A.8}
\]

The collinear contradirectional coupling described here functions as reflector and can have applications such as wavelength filters and dividers, and lasers. The reflector is called distributed Bragg reflector or Bragg grating.
respectively, similar to the coupled-mode treatment of Bragg grating structures.

Thus, the local electric and magnetic fields of the propagating modes in wide (“+”) and narrow (“−”) parts of the grating can be expressed as:

\[ E_\pm = A_\pm^\mu E_\pm^\mu + A_\pm^\nu E_\pm^\nu + E_\pm^r \]  
\[ H_\pm = A_\pm^\mu H_\pm^\mu + A_\pm^\nu H_\pm^\nu + H_\pm^r \]  

where \( A_\pm^\mu \) and \( A_\pm^\nu \) are the amplitudes for the forward and backward local normal modes, respectively, and \( E_\pm^r \) and \( H_\pm^r \) stand for the electric and magnetic fields of the radiation modes, respectively. In such a case, a transfer matrix can be defined to relate amplitudes of the local modes of the “+” sections to the “−” ones:

\[
\begin{pmatrix}
A_+^\mu \\
A_+^\nu
\end{pmatrix}
= T_+ \begin{pmatrix}
A_-^\mu \\
A_-^\nu
\end{pmatrix}
\]  

where:

\[ T_\pm = \begin{pmatrix}
t_+ & t_- \\
t_- & t_+
\end{pmatrix}\]

\[ t_\pm = \frac{1}{4} \iint \left[ E_+^\mu \times H_+^\mu \pm E_+^\nu \times H_+^\nu \right] dx \, dy \]

A transfer matrix accounting for the propagation of the local modes inside “+” and “−” sections can be defined as:

\[ P_\pm = \begin{pmatrix}
e^{-j \gamma_\pm A_\pm} & 0 \\
0 & e^{j \gamma_\pm A_\pm}
\end{pmatrix}\]

where \( \gamma_\pm \) stands for the complex propagation constants of the optical modes (in lossless case, \( \gamma_\pm = \beta_\pm \)). The transfer matrix for a unit cell (from a “+” section to the next “+” section) can thus be written as:

\[ T_{++} = T_{+-} P_- T_{-+} P_+ \]

where \( T_{-+} = T_{+-}^{-1} \). Thus, assuming that the grating consists of \( N \) periods (\( L = N \Lambda \)), the transfer matrix for the whole structure can be written as \( T_{++}^N \).
If one solves the equations for the transfer matrix method, the reflection at the input end of the Bragg structure can be expressed as eq. A.16. Readers are referred to [187] for more details.

\[
\begin{align*}
R_{tmm} &= \left| \frac{\sinh(NAQ_{+g}) e^{-j\alpha_2}}{\cosh(NAQ_{+g} - j\alpha_2)} \right|^2 \\
\end{align*}
\]

where:

\[
\begin{align*}
\alpha_2 &= \gamma_+\Lambda_+ - \frac{\pi}{2} \\
\sin \alpha_+ &= \frac{S_{+2}}{S_{+3}} \\
Q_{+g} &= j\frac{\gamma_{+2} - \gamma_{+1}}{2} \\
e^{-j\gamma_{+1}\Lambda} &= -S_{+1} - S_{+3}\cos \alpha_+ \\
e^{-j\gamma_{+1}\Lambda} &= -S_{+1} + S_{+3}\cos \alpha_+ \\
\end{align*}
\]

and:

\[
\begin{align*}
S_{+1} &= -t_+^2 \cos(\gamma_+\Lambda_+ + \gamma_-\Lambda_-) + t_-^2 \cos(\gamma_+\Lambda_+ - \gamma_-\Lambda_-) \\
S_{+2} &= +t_+^2 \sin(\gamma_+\Lambda_+ + \gamma_-\Lambda_-) - t_-^2 \sin(\gamma_+\Lambda_+ - \gamma_-\Lambda_-) \\
S_{+3} &= 2t_+ t_- \sin(\gamma_-\Lambda_-) \\
\end{align*}
\]

As suggested by eqs. A.16 and A.17, the reflection \( R_{tmm} \) is maximized if \( \Re(\gamma_+\Lambda_+) = \pi/2 \). A similar condition holds for the “−” section: the condition for maximized reflection is \( \Re(\gamma_-\Lambda_-) = \pi/2 \).

It can be shown that if small perturbation assumptions are made, i.e., if the Bragg gratings do not significantly affect performance characteristics of the waveguide such as its effective index, the results obtained from TMM will match those from coupled-mode treatment of Bragg reflectors [187].
Appendix B

Modal Analysis of SHG in LN nanopillars

The experimental procedure for investigation of SHG in LN nanopillars is fully described in **Paper A**. In section 3.4, an introductory analysis of SHG inside a guiding media is provided. Here, a more detailed theoretical analysis for the SHG process will be represented. Modal properties of all guided modes at FF and SH frequencies are first simulated by COMSOL Multiphysics software, while their theoretical overlap integrals are calculated using MATLAB software. These simulated insights have enabled us to calculate a theoretical Figure-of-Merit (FOM) measuring relative contribution of longitudinally-driven SHG, explained later.

The nanopillars investigated in **Paper A** have been simulated to be single-mode in the FF wavelength of 850 nm (see **Paper A** for more details). Furthermore, since performed polarimetric experiments have only concerned X-polarized component of the generated SH signal, here I focus only on this component. The X-polarized SH signal can be expressed as:

\[
P_{X}^{(SH)} = -2\varepsilon_0 d_{22} E_{X}^{(FF)} E_{Y}^{(FF)} + 2\varepsilon_0 d_{31} E_{X}^{(FF)} E_{Z}^{(FF)}
\]

6B.1

The first term in the right-hand side of eq.6B.1 represents a *purely transverse* contribution to the SHG: it arises only from transverse components of the guided modes. If we describe the corresponding interaction as \(E_X^{(FF)} + E_Y^{(FF)} \rightarrow SH_\nu\), where \(SH_\nu\) is the generated and guided SH mode, the coupling coefficient described in eq. 3.34 can be rewritten as:

\[
\kappa_{\nu XY} = \frac{d_{22} \omega_{SH}}{\pi c N_\nu} \frac{\iint_{\text{pillar}} E_X(x, y) E_Y(x, y) E_{X\nu}(x, y) dx dy}{\iint E_{Y\nu}^2(x, y) dx dy}
\]

6B.2

where \(E_X(x, y) = \hat{x} \cdot E_{HE_{11}}^{(X)}\) and \(E_Y(x, y) = \hat{y} \cdot E_{HE_{11}}^{(Y)}\) are the transverse profiles of the relevant FF modes, and \(E_{X\nu}(x, y) = \hat{x} \cdot E_\nu\) is the X-polarized component of the SH mode \(\nu\).
The second term in the right-hand side of eq. B.1 corresponds to additional SHG contributions, involving transverse ($E_x$) and longitudinal ($E_z$) components of the guided FF mode. Similar to the previous discussions, the coupling coefficient can be expressed as:

$$\kappa_{vZ} = \frac{d_{31} \omega_{SH}}{\pi c N_v} \frac{\iint \text{pillar} E_x(x,y) E_z(x,y) E_{Xv}(x,y) dx \ dy}{\iint E_z^2(x,y) dx \ dy} \tag{B.3}$$

where $E_z(x,y) = \hat{z} \cdot E_{HE_{11}}$ is longitudinal component of the guided FF mode.

Symmetric arguments and numerical computations of the SHG coupling coefficients of eqs. 6B.2 and B.3 for the whole spectrum of SH modes supported by the nanopillar waveguides (Figure 1(c) in paper A), allow us to significantly restrict the set of interactions to be considered in the analysis to a subset of SHG processes, listed in Table I in Paper A.

Consequently, the total $X$-polarized component of the SH signal is proportional to:

$$P^{(SH)}_x(L) \propto |E_{X0}|^2 \left[ |E_{Y0}|^2 \sum_{v=1}^{3} \kappa_v^2 \sin c^2 \left( \frac{\Delta \beta_v L}{2} \right) 
+ |E_{X0}|^2 \sum_{v=4}^{5} \kappa_v^2 \sin c^2 \left( \frac{\Delta \beta_v L}{2} \right) \right] \tag{B.4}$$

where $E_{X0} = A_X(0)$ and $E_{Y0} = A_Y(0)$ are the slowly varying amplitudes of the guided FF modes, $HE_{11}^{(X)}$ and $HE_{11}^{(Y)}$, respectively, and $\kappa_v$ and $\Delta \beta_v$ stand for coupling efficiency and phase mismatch for each SHG interaction (illustrated as $m$ indices in Table I, Paper A).

Thus, one can define coefficients $\eta_a$ and $\eta_b$, describing the efficiencies of $X$-polarized SHG in each nanopillar, according to the first and second terms in the right-hand side of eq. B.4, respectively. A theoretical figure-of-merit (FOM) can then be defined as $FOM_{(TH)} = \sqrt{\eta_b / \eta_a}$ illustrating relative contribution of longitudinal term to the overall SHG process. The experimental values of FOM, described in Paper A, are obtained through fitting curves into experimental results.
References


REFERENCES


REFERENCES


REFERENCES


REFERENCES


REFERENCES


REFERENCES


REFERENCES


REFERENCES


