MgO:LiNbO₃ NONLINEAR WAVELENGTH CONVERTERS — FROM VISIBLE TO MID-INFRARED

BY

JIAN SUN

M.A.Sc

MgO:LiNbO₃ NONLINEAR WAVELENGTH CONVERTERS — FROM VISIBLE TO MID-INFRARED

MgO:LiNbO₃ NONLINEAR WAVELENGTH CONVERTERS — FROM VISIBLE TO MID-INFRARED

By

Jian Sun

M.A.Sc

A Thesis

Submitted to the School of Graduate Studies

in Partial Fulfillment of the Requirements

for the Degree

Doctor of Philosophy

McMaster University © Copyright by Jian Sun, 2014

Master of Applied Science (2014)

McMaster University

(Engineering Physics)

Hamilton, Ontario

- i -

TITLE:MgO:LiNbO3 Nonlinear Wavelength
Converters – From Visible to Mid-InfraredAUTHOR:Jian Sun
M. A. Sc. (McMaster University)SUPERVISOR:Dr. Chang-qing Xu
Professor, Department of Engineering
Physics

COMMITTEE MEMBER:

Dr. Kalaichelvi Saravanamuttu Associate Professor, Department of Chemistry

Dr. Qiyin Fang

Associate Professor, Department of

Engineering Physics

NUMBER OF PAGES: xvii, 116

Abstract

Nonlinear optics studies high-order interactions between light and matter. By exploiting nonlinear optical effects, scientists are able to construct light sources with new wavelengths and functionalities. From the deep ultraviolet (UV) to the far infrared, laser wavelengths that are unobtainable from an active gain medium can be achieved through wavelength conversion in nonlinear optical crystals. Although nonlinear optical effects are only significant under intense light fields, the development of laser technologies over the past few decades has made it a practical method in modern laboratories and even consumer products.

Since the first proposal of periodically structured nonlinear materials in the early 1960s, quasi-phase matching (QPM) has become one of the major methods to achieve high efficiency nonlinear optical conversion. MgO doped periodically poled LiNbO₃ (MgO:PPLN) possesses a large second-order nonlinear coefficient (d₃₃), a wide transparency window from near-UV to mid-infrared (mid-IR), and elevated resistance to photon induced damage under high light power. It has been widely used in second-order nonlinear optical interactions such as second harmonic generation (SHG), sum frequency generation (SFG), and difference frequency generation (DFG). More importantly, high-quality optical waveguides can be formed on MgO:PPLN crystal surfaces, making it attractive for integrated photonics devices or fiber-based optical systems.

In this study, we developed state-of-the-art fabrication techniques for creating MgO:PPLN ridge waveguides. By combining the annealed proton-exchange (APE) process and precise machining, MgO:PPLN ridge waveguides were fabricated. The waveguides exhibited good uniformity over 5cm length, and a measured surface roughness less than 4nm. The measured propagation loss at near-IR wavelength was

about 0.2dB/cm. The optical properties of the waveguides can be tuned by changing the APE conditions and precise machining parameters. The waveguides can be optimized for different applications across its transparency spectrum. Compared to other types of waveguides, such as buried APE channel waveguides or wafer-bonded waveguides, our fabrication method focuses on flexibility, repeatability, high efficiency, and low cost, which are important for practical applications.

Based on literature reports and our experiment results, a simulation model of the APE MgO:PPLN waveguides was built. Waveguide characteristics such as refractive index profile, mode profile and nonlinear conversion efficiency can be calculated. The simulation tool can be used in the design stage to predict the performance of the waveguide, or in the analysis stage to find out possible cause of problems observed in experiments.

To optimize the fabrication conditions of the waveguide, we used the Fourier transform infrared (FTIR) spectroscopy to study the evolution of the H_xLi_{1-x}NbO₃ composite during the proton-exchange (PE) and annealing process. Using the transmission spectrum, we were able to compare the behaviors of APE LiNbO₃ and APE MgO:LiNbO₃ crystals. The diffusion constant of APE MgO:LiNbO₃ in the PE process was calculated through the comparison. The evolution of absorption profiles during annealing revealed three distinguishable absorption peaks at slightly different wavelengths, which indicate that at least three different crystal phases exist in APE MgO:LiNbO₃. Measurement of waveguide propagation loss and phase matching wavelengths showed that significant changes of refractive index and the second-order nonlinear coefficient occurred during annealing. Analysis of the obtained data showed that in order to obtain high quality APE MgO:LiNbO₃ waveguides with stable and repeatable performance, the PE time and annealing time must satisfy a certain

relationship.

Using MgO:PPLN ridge waveguides with optimized performance, we were then able to develop high efficiency nonlinear wavelength converters for 532nm green light generation. Using a 1064nm pump laser, 532nm light was generated through SHG. 398mW of continuous-wave green light output was obtained from a 8.5µm-wide, 1.4cm-long waveguide. Accounting for the Fresnel loss at the waveguide facet, we calculated that a net amount of 466mW of green light was generated in the waveguide. The corresponding optical-to-optical conversion efficiency from 1064nm to 532nm was 69.7%.

An increased insertion loss and heat generation from the ridge waveguide was observed when over 100mW of pump power was injected into the waveguide. This was associated with light induced effects such as green induced infrared absorption (GRIIRA). We characterized the GRIIRA absorption coefficient of MgO:LiNbO₃ waveguides, and compared the results with and without APE. By considering the amount of light absorbed in the waveguides, we simulated the temperature profile under various pump power. Due to the low thermal conductivity of MgO:LiNbO₃, a noticeable temperature gradient was established between the waveguide core and the environment, which in turn affected the device's phase matching condition. Considering this temperature gradient, we performed SHG simulations in the MgO:PPLN crystal, and found that the waveguide performance is significantly limited by the GRIIRA effect. Therefore, thermal management is crucial for MgO:PPLN operating under high power.

We also developed APE MgO:PPLN ridge waveguides for efficient SHG of 1.5µm communication band light. Using a 19mm-long 12µm-wide waveguide, 780nm light was successfully generated using a tunable laser working at 1560nm. The measured power and efficiency performances of the waveguide were promising. The combination of a high

power 1.5µm-band tunable laser and a high efficiency SHG waveguide can be used as a flexible and economic pump source for second-stage nonlinear interactions such as optical parametric generation or photon pair generation.

A mid-IR laser with milliwatt-level output was also developed using the APE MgO:PPLN ridge waveguide. We fabricated 36mm long waveguides with various ridge width and poling periods. The characteristics of the waveguides were tested with spontaneous parametric down conversion (SPDC) spectroscopy. Using a 1W 1064nm pump laser, we obtained the 1550nm-band phase matched SPDC signal, which was strong enough to be measured with a spectrum analyzer. Using the SPDC signal, we were able to determine the phase-matching wavelengths and temperature tuning characteristics of each waveguide. Following the DFG method, we generated 1.6mW of 3.3µm light using a 1063nm laser and a 1554nm dual-wavelength fiber laser. We concluded that a widely tunable mid-IR laser could be built using the ridge waveguide, a 1.06µm-band laser, and a tunable 1.5µm-band laser. Although the performance of the APE MgO:PPLN waveguide in mid-IR generation was limited by several factors such as propagation loss and weak light confinement, the generated milliwatt-level output power is sufficient enough for many spectroscopy and sensing applications.

Acknowledgements

I would like to thank my supervisor Dr. Chang-qing Xu for his continuous support and guidance through my four years of Ph.D. study. To a graduate student, there is nothing more important than having a knowledgeable and easygoing professor. Dr. Xu's profound knowledge and experience, as well as his professional attitude have placed enormous influences on my way of pursuing truth and wisdom.

I also would like to thank my committee members, Dr. Kalaichelvi Saravanamuttu and Dr. Qiyin Fang, for their full support and guidance through my study. Their professional opinions and kind help are invaluable treasures that I will cherish forever.

I would also like to thank all faculty members and staff of the department of Engineering Physics. Without their continuous supports and help, I would not be able to complete my study on time.

Table of Content

Chapter 1. Introduction	1
1-1. Optical Nonlinear Wavelength Conversion	1
1-2. Periodically Poled MgO:LiNbO ₃ Crystal	2
1-3. MgO:PPLN Waveguide for Nonlinear Wavelength Conversion	4
1-4. Motivation of This Study	6
Chapter 2. Theoretical Background	12
2-1. Second Order Nonlinear Optical Interaction	. 12
2-2. Quasi-Phase Matching	. 16
2-3. Annealed Proton-Exchange in LiNbO ₃ Crystal	. 17
2-4. Simulation of APE MgO:LiNbO3 Ridge Waveguide	. 21
2-5. Summary	. 24
Chapter 3. Characterization of APE MgO:LiNbO3 Using FTIR Spectroscopy	25
3-1. Annealed Proton-Exchange in LiNbO3 and MgO:LiNbO3	. 25
3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃3-2. Sample Preparation and Test Conditions	. 25 . 27
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃ 3-2. Sample Preparation and Test Conditions 3-3. Comparison of PE LiNbO₃ and PE MgO:LiNbO₃ 	. 25 . 27 . 28
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃ 3-2. Sample Preparation and Test Conditions	. 25 . 27 . 28 . 32
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃ 3-2. Sample Preparation and Test Conditions	. 25 . 27 . 28 . 32 . 39
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃ 3-2. Sample Preparation and Test Conditions	. 25 . 27 . 28 . 32 . 39 . 44
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃	. 25 . 27 . 28 . 32 . 39 . 44
 3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃	. 25 . 27 . 28 . 32 . 39 . 44 49

4-3. Repeatability of the Dicing Process	53
4-4. Summary	54
Chapter 5. 1064nm to 532nm Second Harmonic Generation	56
5-1. Application Background	56
5-2. Waveguide Design and Fabrication	57
5-3. Green Light Generation using APE MgO:PPLN Ridge Waveguide	60
5-4. Green Induced Infrared Absorption in MgO:LiNbO ₃	63
5-5. Measurement of GRIIRA Coefficient in APE MgO:PPLN	65
5-6. GRIIRA Induced Temperature and Refractive Index Modulations	70
5-7. Influence of the GRIIRA Effect on SHG Performance	77
5-8. Summary	81
Chapter 6. 1.5-µm Communication Band Second Harmonic Generation	86
6-1.Application Background	86
6-2. Device Design and Fabrication	87
6-3. Experiment Result	90
6-4. Summary	93
Chapter 7. 3.3 µm-Band Mid-Infrared Difference Frequency Generation	96
7-1. Application Background	96
7-2. Waveguide design and fabrication	97
7-3. Characterization of Phase Matching Conditions by SPDC Spectroscopy	y 99
7-4. Milliwatt-Level Mid-IR Generation	103
7-5. Summary	107
Chapter 8. Conclusions	109

Appendix	
FTIR spectrum of APE MgO:LiNbO3 crystals with vario	us PE and annealing time
List of Publications	

List of Figures

Figure 2-1	Extraordinary refractive index of 5mol% MgO doped LiNbO3 crystal.		
		18	
Figure 2-2	Calculated PE layer thickness in PE MgO:LiNbO ₃	19	
Figure 2-3	Calculated refractive index of APE MgO:LiNbO3 with 2 hours of H	PΕ	
	time, and 0~6 hours of annealing time.	21	
Figure 2-4	(a) simulated and (b) measured waveguide mode profile of a 6μ m wide	de	
	APE MgO:LiNbO3 ridge waveguide fabricated with 2 hours PE tin	ne	
	and 6 hours of annealing time	22	
Figure 2-5	(a) Horizontal and (b) vertical cross-section mode profiles of result	lts	
	shown in Fig. 2-4.	23	
Figure 3-1	(a) Absorption spectra of none-MgO-doped PE LiNbO ₃ . (b) Measure	ed	
	light transmittance at 3501cm ⁻¹ (solid), and exponential numeric	al	
	fitting (dashed) in PE LiNbO ₃	30	
Figure 3-2	(a) Absorption spectra of PE MgO:LiNbO ₃ . (b) Measured lig	t	
	transmittance at 3501cm ⁻¹ (solid), and exponential numerical fitting	ng	
	(dashed) in PE MgO:LiNbO ₃	31	
Figure 3-3	Absorption spectra of (a) APE LiNbO ₃ and (b) APE MgO:LiNbO ₃ wi	ith	
	4 hours of proton-exchange time and 0~24 hours of annealing time.	34	
Figure 3-4	Normalized absorption spectrum of phase a , b , and c crystals in AF	ΡE	
	MgO:LiNbO ₃	35	

Figure 3-5	-5 Curve fitting result of an APE MgO:LiNbO ₃ sample with 4 hours		
	PE time and 8 hours of annealing. Individual absorption curves from	m	
	different amount of phase <i>a</i> , <i>b</i> , <i>c</i> crystals are shown	35	
Figure 3-6	Amount of phase α crystals in APE LiNbO ₃ during annealing. The P	E	
	time on each sample is 0~24 hours	36	
Figure 3-7	Amount of phase β crystals in APE LiNbO ₃ during annealing. The P	E	
	time on each sample is 0~24 hours	36	
Figure 3-8	Amount of phase <i>a</i> crystals in APE LiNbO ₃ during annealing. The P	Е	
	time on each sample is 0~24 hours	37	
Figure 3-9	Amount of phase b crystals in APE LiNbO ₃ during annealing. The P	Е	
	time on each sample is 0~24 hours	37	
Figure 3-10	Amount of phase c crystals in APE LiNbO ₃ during annealing. The P	Е	
	time on each sample is 0~24 hours	38	
Figure 3-11	X-ray diffraction results of APE MgO:LiNbO3 crystals	38	
Figure 3-12	SHG spectra of the MgO:PPLN slab waveguide with 4 hours of proto	n	
	exchange and 0~24 hours of annealing time	40	
Figure 3-13	(a) Output power of the APE MgO:PPLN slab waveguide. (b) SHe	G	
	signal strength and normalized conversion efficiency of the waveguide	e.	
		41	
Figure 3-14	Relationship between the PE time and the annealing time for AP	E	
	MgO:LiNbO ₃ waveguide to reach phase b depletion and stable	le	
	performance.	44	

Figure 4-1	Fabrication procedures of APE MgO:PPLN crystals. The PE process is		
	carried out at 200 °C; the annealing process is performed at 350 °C.	49	

- Figure 4-2 MgO:PPLN ridge waveguides processed by precise diamond blade dicing. The ridges are about 6μm wide and 30μm high. The waveguide facets are 70μm high.
- Figure 4-3 (a) SEM images of a MgO:LiNbO₃ ridge waveguide. The red square in the lower left part indicates the area of the AFM test. (b) and (c) are the AFM test result. The measured surface roughness RMS value is 3.72 nm.

- Figure 5-3 Experiment setups for 532nm green light SHG using APE MgO:PPLN ridge waveguide. The waveguide chip is flip-mounted to reduce thermal resistance. 61

Figure 5-12 (a) The 3D waveguide model used in the heat transfer simulation. (b)

Cross section view of the model. The red area indicates the waveguide,

which is the heat generating source	73
which is the heat generating source.	

- Figure 5-14 Waveguide core temperature increase caused by the GRIIRA effect.Simulation results in (a) and (b) are based on the results in Fig. 5-10and Fig. 5-11 respectively.75

- Figure 5-18 Simulated temperature distribution profile generated by the GRIIRA effect in MgO:LiNbO₃ waveguide. (a) The crystal is placed on a temperature controller with the waveguide facing upwards. (b) The waveguide is flip-mounted on a temperature controlled silicon substrate with adhesives. 80
- Figure 6-1 Calculated refractive index profile for APE MgO:LiNbO₃ waveguide at (a) 1560nm, and (b). 780nm. The waveguide is processed with 2.5

Figure 6-2	Microscope photos of the fabricated ridge waveguides. The wavegui	ide
	facets are cut to 9° angle to prevent back-reflection. The chip conta	ins
	a 10µm wide and a 12µm ridge waveguide	89

- Figure 6-3 Normalized SHG spectrum of (a) 12µm wide and (b) 10µm wide ridge waveguide. The spectrum is measured using a 1.5µm-band broadband light source.
 91

- Figure 7-2 Experiment setup of the SPDC spectrum test. A high power 1064nm fiber laser is used to couple over 1W of light into the waveguide. The 1550nm-band SPDC signal is collected, filtered, and measured in the spectrum analyzer. 100
- Figure 7-4SPDC spectrum of a12μm wide waveguide with 29.5μm (left) and30μm (right) poling period.101
- Figure 7-5 SPDC spectrum of a $12\mu m$ wide waveguide measured at 25° C (1535nm), 45 °C (1541.6nm), and 65 °C (1550.6nm). 102

	Figure 7-6	Experiment setup	for milliwatt-level mid-IR light generation	104
--	------------	------------------	---	-----

Figure 7-7 1057nm and 1554nm light power coupled into the waveguide. The horizontal axis is the pump current of the dual wavelength fiber laser.

- Figure 7-9 FTIR spectrum of bulk MgO:LiNbO₃ crystal and APE MgO:LiNbO₃ crystal with 12 hours of PE and 12 hours of annealing. The beam direction is perpendicular to the crystal surface with APE. The transmission difference at 3.3µm wavelength is around 0.3%. 107

Chapter 1. Introduction

1-1. Optical Nonlinear Wavelength Conversion

Since P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich first discovered the phenomenon of optical harmonics in 1961 [1-1], nonlinear optics, as an independent branch of modern physics, began to blossom. Soon after the milestone discovery, J. A. Giordmaine [1-2], P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage [1-3] revealed the dispersion relations necessary to obtain maximum second harmonic, known as the phase-matching conditions. In 1962, J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan published the famous paper "Interactions between Light Waves in a Nonlinear Dielectric" [1-4], which laid the theoretical foundation stone of optical nonlinear interactions between light and matter.

50 years later, nonlinear optics is now an extensive research field that completely exceeded the expectations of the pioneers [1-5]. Along with the development of powerful lasers, nonlinear optical devices can be found in many modern laboratories. The applications of nonlinear optical devices include material science, lasers, communications, quantum information, spectroscopy, biomedical imaging, and even consumer electronics.

In laser physics, nonlinear optics has become a powerful tool to obtain new wavelengths. For example, a high power solid-state 532nm green laser can be built with a 1064nm infrared (IR) pump laser and a second harmonic generation (SHG) crystal. A 589nm sodium-line yellow laser can be constructed using two IR pump lasers and a sum frequency generation (SFG) crystal. 3~9µm mid-IR laser can be made using two IR pump lasers and a difference frequency generation (DFG) crystal. Highly efficient crystals such

as potassium titanyl phosphate (KTP), lithium niobate (LiNbO₃), lithium borate (LBO), barium borate (BBO), and silver thiogallate (AgGaS₂) have been well studied for different types of wavelength conversions.

Through nonlinear effects, researchers can realize new functionalities and build new light sources. Super-continuum radiation can be generated by launching femtosecond laser pulses into a photonic crystal fiber [1-6]. Polarization entangled photon pairs can be obtained from spontaneous parametric down conversion (SPDC) in a nonlinear optical crystal [1-7]. The frontiers of nonlinear optics are expanding at an unprecedented speed every day.

1-2. Periodically Poled MgO:LiNbO₃ Crystal

In 1962, J. A. Armstrong et al. proposed that a periodically arranged dielectric with opposite nonlinear coefficients could dramatically enhance the nonlinear conversion efficiency. Later known as quasi-phase matching (QPM), this method was investigated in detail during that period [1-8]-[1-12]. In the QPM method, by periodically reversing an axis of a nonlinear optical crystal, the generated light can be enhanced after constructive interference through propagation.

LiNbO₃ is a ferroelectric crystal with negative uniaxial birefringence. The crystal structure of LiNbO₃ belongs to the trigonal crystal system. The oxygen atoms in LiNbO₃ crystal are close-packed in a distorted hexagonal configuration. The octahedral interstices of the structure are 1/3 filled with Li⁺ ions, 1/3 filled with Nb⁵⁺ ions, and 1/3 vacant. Below the Curie temperature of 1210°C, LiNbO₃ exhibits spontaneous polarization along its z-axis due to the slight displacement of Li⁺ and Nb⁵⁺ ions relative to the oxygen

octahedra [1-13]. The polarization direction of LiNbO₃ can be inversed by applying high-voltage electric pulses along its z-axis.

To realize high efficiency QPM light generation, it is crucial to obtain z-axis periodically poled LiNbO₃ (PPLN). In 1978, S. Miyazawa from NTT Corporation first discovered the ferroelectric domain inversion phenomena on the +z plane of Ti-diffused LiNbO₃ [1-14], which has enabled large area surface periodical poling and effective nonlinear interactions in waveguides [1-15] [1-16]. However, bulk type PPLN was not produced until 1992, when the electric poling method was first demonstrated by M. Yamada *et al.* from Sony Corporation [1-17]. In this method, a metal electrode with a specifically designed period is deposited on the +z surface of a z-cut LiNbO₃ crystal. An electric pulse with a voltage slightly higher than the coercive field of LiNbO₃ is applied to reverse the crystal domain under the electrode. This is now the most widespread technique to make high-quality large-area PPLN crystals.

Other methods of fabricating PPLN have also been investigated by researchers. For example, surface poling can be achieved by Li_2O out-diffusion [1-18] or electron beam bombardment [1-19] [1-20]. Bulk PPLN crystal can be obtained by the laser-heated pedestal growth method [1-21]-[1-23], or the corona discharge method [1-.24]. It also has been found that photon illumination of LiNbO₃ can reduce its coercive field and thus assist the electric poling process [1-25]-[1-27].

LiNbO₃ has a transmission window from 400nm to 5 μ m, which covers a wide range of useful visible and IR wavelengths. Its largest second-order nonlinear coefficient (d₃₃) is 34pm/V, which is much larger than other commonly used nonlinear crystals such as KTP (d₃₃=15pm/V) or LBO (d₃₁=1.1pm/V). LiNbO₃ can be grown via the Czochralski (CZ) process, which dramatically reduces the unit manufacturing cost of each crystal wafer. The combined advantages have made PPLN an ideal candidate for high efficiency, low cost nonlinear optical wavelength conversion devices.

However, the high power performances of LiNbO₃ crystals are limited. Researchers have found that under kW/cm^2 power density, LiNbO₃ is subject to severe photorefractive damage (PRD), which causes self-modulated refractive index change. Another effect is light induced absorption, which increases under higher light intensity. This results in excessive power loss and heat generation, which causes self-induced temperature change in the crystal. These photo-induced phenomena are considered to be linked with the intrinsic and extrinsic defects generated during the CZ growth process.

It has been widely recognized that doping LiNbO₃ with 5mol% MgO can dramatically reduce those photo-induced effects [1-28]-[1-.30]. Today, 5mol% MgO doped LiNbO₃ (MgO:LiNbO₃) and periodically poled MgO:LiNbO₃ (MgO:PPLN) have largely replaced the undoped crystals. Although researchers have found that other dopants such as ZnO [1-31] [1-32] can also effectively suppress photo-induced effects, MgO:PPLN remains the most widely used crystal in the LiNbO₃ family for nonlinear optical interactions.

1-3. MgO:PPLN Waveguide for Nonlinear Wavelength Conversion

Another advantage of LiNbO₃ over other nonlinear optical crystals is the capability to form high quality optical waveguide on its surfaces. In 1974, R. V. Schmidt and I. P. Kaminow produced Ti-diffused optical waveguide on the LiNbO₃ crystal surface [1-33]. Ti atoms were diffused into the crystal under high temperature around 1000 °C. This produced a refractive index increment about 10⁻³. In 1982, J. L. Jackel, C. E. Rice, and J. J. Veselka reported another method to produce optical waveguide by proton-exchange [1-34]. Later, thermal annealing was added after the proton-exchange process [1-35] to optimize the waveguide performance. The refractive index contrast produced by the annealed proton-exchange (APE) method is around 0.01.

By placing the LiNbO₃ crystal in a proton source such as melted benzoic acid, the H⁺ ions replace the Li⁺ ions and form an H_xLi_{1-x}NbO₃ compound on the crystal surface. In the following annealing process, the H⁺ ions diffuse further into the crystal and form a stable light guiding layer of a few micrometers. The compound finally rests in the so-called α phase [1-36], which possess a low H⁺ concentration with a fraction *x*<0.12. Within the wave-guiding layer, the ordinary refractive index decreases while the extraordinary one increases. As a result, the APE LiNbO₃ waveguides guide only the TM modes. The APE method also applies to MgO:LiNbO₃ crystals [1-36].

In the early 2000s, LiNbO₃ ridge waveguides started to attract broad attention, because of its better light confinement than buried surface waveguide. Great amount of research efforts have been devoted to fabricating high-quality ridge waveguides either by wet chemical etching [1-37], reactive ion etching [1-38], and wafer bonding plus precise machining [1-39] [1-40]. Wafer-bonded MgO:PPLN ridge waveguides have been successfully used in generating UV [1-41], blue [1-.42], green [1-43], and mid-IR light [1-44] with high conversion efficiency. The fabrication method to produce ridge waveguides by wafer-bonding is very complicated. First, an MgO:PPLN wafer is brought into contact with a LiTaO₃ wafer using adhesive or after plasma activation. Second, after thermal treatment, the bonded MgO:PPLN wafer is lapped and polished to reduce its thickness. The remaining MgO:PPLN thin film is typically around 5~25µm depending on

the application. Finally, the crystal film is diced into ridge waveguides by the precise diamond blade dicing technique. As a result, the cost of each waveguide is noticeably higher than the ion doped or proton-exchanged waveguide.

1-4. Motivation of This Study

For practical applications, it is necessary to develop a device with high efficiency and best cost-performance ratio. It is also crucial to find out a fabrication process that is repeatable and reliable. In this study, we focus on developing novel design and fabrication techniques for APE MgO:PPLN ridge waveguides with stable performance and low cost. Using the waveguides, we also aim to demonstrate highly efficient nonlinear wavelength conversion on 1064nm to 532nm green light generation. Photon-induced effects and absorptions, which present negative effects on wavelength conversions, are also studied. From the obtained results, we can conclude that the APE MgO:PPLN waveguides are excellent candidates for nonlinear wavelength conversion with milliwatt to hundred milliwatt level output power.

REFERENCES:

- [1-1]. P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, "Generation of Optical Harmonics", Phys. Rev. Lett. 7, 118 (1961).
- [1-.2]. J. A. Giordmaine, "Mixing of Light Beams in Crystals", Phys. Rev. Lett. 8, 19

(1962).

- [1-3]. P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, "Effects of Dispersion and Focusing on the Production of Optical Harmonics", Phys. Rev. Lett. 8, 21 (1962).
- [1-4]. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions between Light Waves in a Nonlinear Dielectric", Phys. Rev. 127, 1918 (1962).
- [1-5]. N. Bloembergen, "Nonlinear Optics: Past, Present, and Future", IEEE J. Selec. Topics in Quant. Eletron. 6, 876 (2000).
- [1-6]. J. M. Dudley, G. Genty, and S. Coen, "Supercontinuum generation in photonic crystal fiber", Rev. Mod. Phys. 78, 1135 (2006).
- [1-7]. P. G. Kwiat, K. Mattle, H. Weinfurter, and A. Zeilinger, "New High-Intensity Source of Polarization-Entangled Photon Pairs", Phys. Rev. Lett. 75, 4337 (1995).
- [1-8]. P. A. Franken and J. F. Ward, "Optical harmonics and nonlinear phenomena", Rev. Mod. Phys. 35, 23 (1963).
- [1-.9]. R. C. Miller, D. A. Kleinman, and A. Savage, "Quantitative studies of optical harmonic generation in CdS, BaTiO₃, and KH₂PO₄ type crystals", Phys. Rev. Lett. 11, 146 (1963).
- [1-10]. N. Bloembergen and A. J. Sievers, "Nonlinear optical properties of periodic laminar structures", Appl. Phys. Lett. 17, 483 (1970).
- [1-11]. S. Somekh, and A. Yariv, "Phase matching by periodic modulation of the nonlinear optical properties", Opt. Commun. 6, 301 (1972).

- [1-12]. J. D. McMullen, "Optical parametric interactions in isotropic materials using a phase-corrected stack of nonlinear dielectric plates", J. Appl. Phys. 46, 3076 (1975).
- [1-13]. R. S. Weis and T. K. Gaylord, "Lithium niobate: summary of physical properties and crystal structure", Appl. Phys. A 37, 191 (1985).
- [1-14]. S. Miyazawa, "Ferroelectric domain inversion in Ti-diffused LiNbO₃ optical waveguide", J. Appl. Phys. 50, 4599 (1979).
- [1-15]. E. J. Lim, M. M. Fejer, and R. L. Byer, "Second-harmonic generation of green light in periodically poled planar lithium niobate waveguide," Electron. Lett. 25, 174 (1989).
- [1-16]. E. J. Lim, M. M. Fejer, R. L. Byer, and W. J. Kozlovsky, "Blue light generation by frequency doubling in periodically poled lithium niobate channel waveguide", Electron. Lett. 25, 731 (1989).
- [1-17]. M. Yamada, N. Nada, M. Saitoh, and K. Watanabe, "First-order quasi-phase matched LiNbO₃ waveguide periodically poled by applying an external field for efficient blue second-harmonic generation", Appl. Phys. Lett. 62, 435 (1993).
- [1-18]. J. Webjorn, F. Laurell, and G. Arvidsson, "Blue light generated by frequency doubling of laser diode light in a lithium niobate channel waveguide", IEEE Photon. Technol. Lett. 1, 316 (1989).
- [1-19]. P. W. Haycock and P. D. Townsend, "A method of poling LiNbO₃ and LiTaO₃ below Tc", Appl. Phys. Lett. 48, 698 (1986).
- [1-20]. R. W. Keys, A. Loni, R. M. De La Rue, C. N. Ironside, J. H. Marsh, B. J. Luff and P. D. Townsend, "Fabrication of domain reversed gratings for SHG in LiNbO₃

by electron beam bombardment", Electron. Lett. 26, 188 (1990).

- [1-21]. M. M. Fejer, J. L. Nightingale, G. A. Magel, and R. L. Byer, "Laser heated miniature pedestal growth apparatus for single crystal optical fibers", Rev. Sci. Instrum. 55, 1791 (1984).
- [1-22]. G. A. Magel, M. M. Fejer, and R. L Byer, "Quasi-phase-matched second-harmonic generation of blue light in periodically poled LiNbO₃", Appl. Phys. Lett. 56, 108 (1990).
- [1-23]. D. H. Jundt, G. A. Magel, M. M. Fejer, and R. L. Byer, "Periodically poled LiNbO₃ for high-efficiency second-harmonic generation", Appl. Phys. Lett. 59, 2657 (1991).
- [1-24]. A. Harada and Y. Nihei, "Bulk periodically poled MgO-LiNbO₃ by corona discharge method", Appl. Phys. Lett. 69, 2629 (1996).
- [1-25]. M. Fujimura, T. Sohmura and T. Suhara, "Fabrication of domain-inverted gratings in MgO:LiNbO₃ by applying voltage under ultraviolet irradiation through photomask at room temperature", Electron. Lett. 39, 719 (2003).
- [1-26]. M. Müller, E. Soergel, and K. Buse, "Influence of ultraviolet illumination on the poling characteristics of lithium niobate crystals", Appl. Phys. Lett. 83, 1824 (2003).
- [1-27]. V. Dierolf and C. Sandmann, "Direct-write method for domain inversion patterns in LiNbO₃", Appl. Phys. Lett. 84, 3987 (2004).
- [1-28]. Y. Furukawa, K. Kitamura, S. Takekawa, A. Miyamoto, M. Terao, and N. Suda, "Photorefraction in LiNbO₃ as a function of [Li]/[Nb] and MgO concentrations",

Appl. Phys. Lett. 77, 2494 (2000).

- [1-29]. Y. Furukawa, K. Kitamura, A. Alexandrovski, R. K. Route, M. M. Fejer, and G. Foulon, "Green-induced infrared absorption in MgO doped LiNbO₃", Appl. Phys. Lett. 78, 1970 (2001).
- [1-30]. M. Asobe, O. Tadanaga, T. Yanagawa, H. Itoh, and H. Suzuki, "Reducing photorefractive effect in periodically poled ZnO- and MgO-doped LiNbO₃ wavelength converters", Appl. Phys. Lett. 78, 3163 (2001).
- [1-31]. T. Volk, N. Rubinina, and M. Wöhlecke, "Optical-damage-resistant impurities in lithium niobate," JOSA B 11, 1681 (1994).
- [1-32]. T. R. Volk, V. I. Pryalkin, and N. M. Rubinina, "Optical-damage-resistant LiNbO₃:Zn crystal," Opt. Lett. 15, 996 (1990).
- [1-33]. R. V. Schmidt and I. P. Kaminow, "Metal-diffused optical waveguides in LiNbO₃," Appl. Phys. Lett. 25, 458 (1974).
- [1-34]. J. L. Jackel, C. E. Rice, and J. J. Veselka, "Proton exchange for high-index waveguides in LiNbO₃," Appl. Phys. Lett. 41, 607 (1982).
- [1-35]. Retzon Chen and C. S. Tsai, "Thermally annealed single-mode proton-exchanged channel-waveguide cutoff modulator," Opt. Lett. 11, 546 (1986).
- [1-36]. J. L. Jackel, "Proton exchange in MgO-doped LiNbO₃", Electron. Lett. 21, 509 (1985).
- [1-37]. H. Hu, R. Ricken, W. Sohler, and R. B. Wehrspohn, "Lithium niobate ridge waveguides fabricated by wet etching," IEEE Photon. Technol. Lett. 19, 417 (2007).

- [1-38]. H. Hu, A. P. Milenin, R. B. Wehrspohn, H. Hermann, and W. Sohler, "Plasma etching of proton-exchanged lithium niobate", J. Vac. Sci. Technol. A 24, 1012 (2006).
- [1-39]. S. Kurimura, Y. Kato, M. Maruyama, Y. Usui, and H. Nakajima,
 "Quasi-phase-matched adhered ridge waveguide in LiNbO₃", Appl. Phys. Lett 89, 191123, (2006).
- [1-40]. Y. Nishida, H. Miyazawa, M. Asobe, O. Tadanaga, H. Suzuki, "0-dB Wavelength Conversion using direct-bonded QPM-Zn:LiNbO₃ ridge waveguide", IEEE Photon. Technol. Lett., 17, 1049 (2005).
- [1-41]. K. Mizuuchi, T. Sugita, K. Yamamoto, T. Kawaguchi, T. Yoshino, and M. Imaeda, "Efficient 340-nm light generation by a ridge-type waveguide in a first-order periodically poled MgO:LiNbO₃", Opt. Lett. 28, 1344 (2003).
- [1-42]. M. Iwai, T. Yoshino, S. Yamaguchi, M. Imaeda, N. Pavel, I. Shoji, and T. Taira, "High-power blue generation from a periodically poled MgO:LiNbO₃ ridge-type waveguide by frequency doubling of a diode end-pumped Nd:Y₃Al₅O₁₂ laser", Appl. Phys. Lett. 83, 3659 (2003).
- [1-43]. H. K. Nguyen, M. H. Hu, Y. Li, K. Song, N. J. Visovsky, S. Coleman, and C. E. Zah, "304 mW green light emission by frequency doubling of a high-power 1060nm DBR semiconductor laser diode," Proc. of SPIE 6890, 689001 (2008).
- [1-44]. O. Tadanaga, T. Yanagawa, Y. Nishida, H. Miyazawa, K. Magari, M. Asobe, and H. Suzuki, "Efficient 3-μm difference frequency generation using direct-bonded quasi-phase-matched LiNbO₃ ridge waveguides", Appl. Phys. Lett. 88, 061101 (2006).

Chapter 2. Theoretical Background

2-1. Second Order Nonlinear Optical Interaction

The electromagnetic interactions are governed by the Maxwell equations. In a lossless dielectric (no free-carrier, non-magnetic) media, the equations are:

$$\nabla \times \mathbf{E} = -\frac{\partial}{\partial t} \mathbf{B}$$
(2-1)

$$\nabla \times \mathbf{H} = \frac{\partial}{\partial t} \mathbf{D}$$
(2-2)

$$\nabla \cdot \mathbf{D} = 0 \tag{2-3}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2-4}$$

Where **E** and **B** are the electric field and the magnetic field, **D** and **H** are the electric displacement field and the magnetizing field. The electric displacement in the material can be written as $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$, where **P** is the polarization density of the material. The polarization response of the material contains high order terms, and can be expressed with the Fourier series:

$$\mathbf{P} = \varepsilon_0 [\chi^{(1)} \mathbf{E} + \chi^{(2)} \mathbf{E}^2 + \chi^{(3)} \mathbf{E}^3 + \cdots]$$
(2-5)

The second term $\varepsilon_0 \chi^{(2)} \mathbf{E}^2$ is the second-order nonlinear response. Suppose we have two different waves $\mathbf{E_1}$ and $\mathbf{E_2}$ input into the media, the second-order terms contain $\varepsilon_0 \chi^{(2)} \mathbf{E_1}^2$, $\varepsilon_0 \chi^{(2)} \mathbf{E_2}^2$, and $\varepsilon_0 \chi^{(2)} \mathbf{E_1} \mathbf{E_2}$. The first two terms represent second-order harmonics of $\mathbf{E_1}$ and $\mathbf{E_2}$, and the third term represents sum or difference frequency generation. In fact, second-order harmonic is one special occasion of sum-frequency generation, in which $\mathbf{E_1}$ equals to **E**₂. Thus, the term $\varepsilon_0 \chi^{(2)} \mathbf{E}_1 \mathbf{E}_2$ is of special interest to us.

In general, the second-order nonlinear optical interaction involves two input waves $\mathbf{E_1}$ and $\mathbf{E_2}$. The interactions generates a third wave $\mathbf{E_3}$, which is the result of the term $\varepsilon_0 \chi^{(2)} \mathbf{E_1} \mathbf{E_2}$. As a result, it is named three-wave interactions. We consider only the second-order three-wave interactions in this study.

Assuming that at the beginning of the interaction, the generated wave \mathbf{E}_3 is negligible, and we consider the second-order polarization density $\varepsilon_0 \chi^{(2)} \mathbf{E}_1 \mathbf{E}_2$, \mathbf{E}_3 can be obtained by solving the Maxwell equations.

For simplicity, we denote:

$$\mathbf{P} = \varepsilon_0 \chi^{(1)} \mathbf{E} + \mathbf{P}_{NL} \tag{2-6}$$

Plug **P** into Eq. 2-2, and replace term $\varepsilon = \varepsilon_0 (1 + \chi^{(1)})$, we obtain:

$$\nabla \times \mathbf{H} = \varepsilon \frac{\partial}{\partial t} \mathbf{E} + \frac{\partial}{\partial t} \mathbf{P}_{NL}$$
(2-7)

From Eq. 2-1, we obtain:

$$\nabla \times \nabla \times \mathbf{E} = -\frac{\partial}{\partial t} \nabla \times \mathbf{B} = -\mu_0 \frac{\partial}{\partial t} \nabla \times \mathbf{H}$$
$$= -\mu_0 \frac{\partial}{\partial t} \left(\varepsilon \frac{\partial}{\partial t} \mathbf{E} + \frac{\partial}{\partial t} \mathbf{P}_{NL} \right)$$
(2-8)

Considering $\nabla \times \nabla \times \mathbf{E} = \nabla (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$, and $\nabla \cdot \mathbf{E} = 0$ in the media, we obtain:

$$\nabla^{2}\mathbf{E} - \mu_{0}\varepsilon \frac{\partial^{2}}{\partial t^{2}}\mathbf{E} = \mu_{0} \frac{\partial^{2}}{\partial t^{2}}\mathbf{P}_{NL}$$
(2-9)

Considering E_3 , we have:

$$\nabla^{2} \mathbf{E}_{3} - \mu_{0} \varepsilon \frac{\partial^{2}}{\partial t^{2}} \mathbf{E}_{3} = \mu_{0} \varepsilon_{0} \chi^{(2)} \frac{\partial^{2}}{\partial t^{2}} \mathbf{E}_{1} \mathbf{E}_{2}$$
(2-10)

Note that $\chi^{(2)}$ is in nature a tensor. However, in a specific situation when the status of **E**₁ and **E**₂, and the orientation of the nonlinear media are chosen, $\chi^{(2)}$ can take a certain value. For example, in PPLN, the incident light is polarized along the z-axis of the crystal, and the generated light is also z-polarized. We have $\chi^{(2)} = d_{33}$ in this particular interaction.

Assume that the incident beams are plane waves travelling along the z direction, we can write the vector fields in scalar expression as:

$$E_{1}(z,t) = E_{1}(z)\exp(ik_{1}z - i\omega_{1}t)$$
(2-11)

$$E_{2}(z,t) = E_{2}(z)\exp(ik_{2}z - i\omega_{2}t)$$
(2-12)

The generated wave E_3 is also a plane wave:

$$E_{3}(z,t) = E_{3}(z)\exp(ik_{3}z - i\omega_{3}t)$$
(2-13)

The frequency $\omega_3 = \omega_1 + \omega_2$.

Plug the above expressions into Eq.2-10, and assume the slowly varying amplitude approximation for continuous-wave light $\frac{d^2}{dz^2}E_3(z) = 0$, we obtain:

$$\frac{d}{dz}E_{3}(z) = \frac{i\omega_{3}d_{33}}{2n_{3}c_{0}}E_{1}(z)E_{2}(z)\exp(i\Delta kz)$$
(2-14)

Ph. D. Thesis – Jian Sun – Engineering Physics McMaster University

In this equation, $\Delta k = k_1 + k_2 - k_3$ represents the wave vector mismatch among the three waves. E₃ represents sum-frequency generation $\omega_3 = \omega_1 + \omega_2$. If we assume E₃ and E₂ are known, and solve for E₁, we obtain difference frequency generation $\omega_1 = \omega_3 - \omega_2$.

$$\frac{d}{dz}E_{1}(z) = \frac{i\omega_{1}d_{33}}{2n_{1}c_{0}}E_{2}^{*}(z)E_{3}(z)\exp(-i\Delta kz)$$
(2-15)

Similarly, we solve for E₂:

$$\frac{d}{dz}E_2(z) = \frac{i\omega_2 d_{33}}{2n_2 c_0} E_1^*(z)E_3(z)\exp(-i\Delta kz)$$
(2-16)

In practice, we use the 1st order finite difference method to solve Eq. 2-14~2-16, and program a computer to simulate the three wave interactions. For example, by dividing the interaction length z into small fragments, we can calculate changes to the field E₃ using Eq. 2-14:

$$dE_{3}(z) = \frac{i\omega_{3}d_{33}}{2n_{3}c_{0}}E_{1}(z)E_{2}(z)\exp(i\Delta kz)\cdot dz$$
(2-17)

The step change dE_3 is then added to E_3 to obtain a new value.

$$E_3(z+dz) = E_3(z) + dE_3(z)$$
(2-18)

Similar calculations can be applied to Eq. 2-15 and Eq. 2-16 to find $E_1(z+dz)$ and $E_2(z+dz)$. The new E₁, E₂, and E₃ values are used in the next step of calculation.

2-2. Quasi-Phase Matching

Considering Eq. 2-14, under the non-depletion conditions, E_1 and E_2 are treated as constants. After integration, we obtain:

$$E_{3}(z) = \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}E_{2}e^{i\Delta kz}$$
(2-19)

Taking the definite integral value from 0 to z in Eq. 2-19, we obtain the E_3 field after the beams propagate from 0 to z:

$$E_{3}(z) = \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}E_{2}e^{i\Delta kz} |_{0}^{z} = \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}E_{2}\left(e^{i\Delta kz} - 1\right)$$

$$= \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}E_{2}\left[\cos(\Delta kz) + i\sin(\Delta kz) - 1\right]$$

$$= \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}E_{2} \cdot 2i\sin\left(\frac{\Delta kz}{2}\right) \cdot \left[\cos\left(\frac{\Delta kz}{2}\right) + i\sin\left(\frac{\Delta kz}{2}\right)\right]$$

$$= \frac{i\omega_{3}d_{33}}{n_{3}c_{0}\Delta k} E_{1}E_{2} \cdot \sin\left(\frac{\Delta kz}{2}\right) \cdot e^{\frac{i\Delta kz}{2}}$$

(2-20)

The intensity can be calculated as:

$$I_{3} = \frac{1}{2} n_{3} c_{0} \varepsilon_{0} E_{3} E_{3}^{*} = \frac{\varepsilon_{0} \omega_{3}^{2} d_{33}^{2}}{2n_{3} c_{0} \Delta k^{2}} E_{1}^{2} E_{2}^{2} \cdot \sin^{2} \left(\frac{\Delta kz}{2}\right)$$
(2-21)

Eq. 2-21 shows that the intensity of the generated wave oscillates along the propagation direction. I₃ increases between 0 to $\frac{\pi}{\Delta k}$, and decreases between $\frac{\pi}{\Delta k}$ to $\frac{2\pi}{\Delta k}$. The result comes from constructive or destructive interference of dE_3 generated from each dz section.

We are able to turn-around the destructive interference by reversing the sign of d_{33}

after each $\frac{\pi}{\Delta k}$ crystal length. This can be realized by reversing the z-axis in the LiNbO₃ crystal. As a result, $\Lambda = \frac{2\pi}{\Delta k}$ is the poling period of the PPLN crystal.

After periodical poling, we obtain a modulated d₃₃ coefficient as follows:

$$d_{33} = \begin{cases} +d_{33}, & n\Lambda < z < (n+1/2)\Lambda \\ -d_{33}, & (n+1/2)\Lambda < z < (n+1)\Lambda \end{cases} n \ge 0, n \text{ is integer} \qquad (2-22)$$

Using Eq. 2-22 and Eq. 2-14~2-16, we are able to simulate the second-order quasi-phase matched nonlinear interactions. By numerically integrating Eq. 2-17, after each $\Lambda/2$, the E₃ field can be calculated as:

$$\Delta E_{3}^{2n+1} = \frac{i\omega_{3}d_{33}}{2n_{3}c_{0}} E_{1}^{2n} E_{2}^{2n} \int_{n\Lambda}^{(n+1/2)\Lambda} e^{i\Delta kz} dz$$

$$= \frac{\omega_{3}d_{33}}{2n_{3}c_{0}\Delta k} E_{1}^{2n} E_{2}^{2n} \left[\exp\left(i\Delta k(n+\frac{1}{2})\Lambda\right) - \exp\left(i\Delta kn\Lambda\right) \right]$$
(2-23)

or:

$$\Delta E_{3}^{2n} = \frac{-\omega_{3} d_{33}}{2n_{3}c_{0}\Delta k} E_{1}^{2n-1} E_{2}^{2n-1} \left[\exp(i\Delta kn\Lambda) - \exp\left(i\Delta k(n-\frac{1}{2})\Lambda\right) \right]$$
(2-24)

depending on the sign of d_{33} coefficient as shown in Eq. 2-22. In Eq. 2-23 and 2-24, *n* represents the spatial series counted in the number of poling periods. By periodically reversing the sign of d_{33} , the SHG wave keeps growing under constructive interference.

2-3. Annealed Proton-Exchange in LiNbO₃ Crystal

The extraordinary refractive index of 5mol% MgO:LiNbO₃ can be calculated as [2-1]:
$$n_e^2 = 5.756 + 2.86 \times 10^{-6} f + \frac{0.0983 + 4.7 \times 10^{-8} f}{\lambda^2 - (0.2020 + 6.113 \times 10^{-8} f)^2} + \frac{189.32 + 1.516 \times 10^{-4} f}{\lambda^2 - 12.52^2} - 0.0132\lambda^2$$

$$(2-25)$$

where f = (T - 24.5)(T + 570.82), T is the temperature in unit of Celsius, and λ is the wavelength in unit of microns. Fig. 2-1 shows the refractive index curve from 400nm to 4 μ m.



Fig. 2-1. Extraordinary refractive index of 5mol% MgO doped LiNbO3 crystal.

After proton-exchange, a layer of $H_x Li_{1-x} NbO_3$ with a step-like index profile is formed on the surface of the crystal. The PE layer thickness can be calculated as:

$$d = 2\sqrt{D(T) \cdot t} \tag{2-26}$$

where t is the PE time, and D(T) is the temperature-dependent diffusion coefficient [2-2].

From the experiments in Chapter 3, the diffusion coefficient in MgO:LiNbO₃ at 200 °C is calculated as $D(200°C)=0.062\mu m^2/h$. Fig. 2-2 shows the calculated PE layer thickness at different exchange time.



Fig. 2-2. Calculated PE layer thickness in PE MgO:LiNbO₃.

In none-doped LiNbO₃, the PE layer mainly contains β phase H_xLi_{1-x}NbO₃. The refractive index difference between the PE layer and the crystal substrate can be calculated as [2-2]:

$$\Delta n_e(\lambda) = \sqrt{a_1 + \frac{a_2}{\lambda^2 - a_3^2}}$$
(2-27)

where a_1 , a_2 , a_3 are 7.43×10^{-3} , 2.64×10^{-3} , and 0.336 respectively. As described inn Chapter 3, we find that the spectrum signatures of the PE layers in MgO:LiNbO₃ and none-doped LiNbO₃ are the same. We can expect the PE layer in MgO:LiNbO₃ is consisted of the same type of compound as that in none-doped LiNbO₃. As a result, Eq.

2-27 should also apply to PE MgO:LiNbO₃.

After annealing, the hydrogen distribution profile as a function of diffusion depth and annealing time can be numerically calculated with the method described by M. L. Bortz and M. M. Fejer [2-2]. The problem can be modeled as a time-dependent *1-D* diffusion problem described by:

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial}{\partial x} \left(D(x,t) \cdot \frac{\partial C(x,t)}{\partial x} \right)$$
(2-27)

where C(x,t) is the normalized hydrogen concentration profile, and D(x,t) is the concentration-dependent diffusion coefficient:

$$D(x,t) = D_0\{a + (1-a)\exp[-b \cdot C(x,t)]\}$$
(2-28)

 D_0 is the asymptotic diffusion coefficient at low proton concentration, and *a* and *b* are constants.

Since both D(x,t) and C(x,t) are space and time dependent, for simplicity, we take the Forward-Time Central-Space (FTCS) scheme to solve Eq.2-27 numerically.

Eq. 2-27 can be written as:

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial D(x,t)}{\partial x} \cdot \frac{\partial C(x,t)}{\partial x} + D(x,t) \cdot \frac{\partial^2 C(x,t)}{\partial x^2}$$
(2-29)

Take the FTCS scheme on Eq.2-29, we obtain:

$$\frac{C_i^{j+1} - C_i^j}{\Delta t} = \frac{D_{i+1}^j - D_i^j}{\Delta x} \cdot \frac{C_{i+1}^j - C_i^j}{\Delta x} + D_i^j \cdot \frac{C_{i+1}^j - 2C_i^j + C_{i-1}^j}{\Delta x^2}$$
(2-30)

where *i* and *j* represents the spatial and time series respectively.

After solving the hydrogen distribution profile, the refractive index profile can be

modeled as [2-2]:

$$n_{APE}(\lambda, x, t) = n_e + 1.06\Delta n_e(\lambda) \cdot C(x, t)$$
(2-31)

After obtaining the refractive index profile, the optical modes in a waveguide can be calculated using a simulation program. Through numerical fitting using parameters a=0.1, b=12 [2-2] in Eq. 2-28, we find the asymptotic diffusion coefficient $D_0=2.6\mu m^2/h$ can provide best fittings for APE MgO:LiNbO₃ waveguides annealed at 350°C.

2-4. Simulation of APE MgO:LiNbO3 Ridge Waveguide

By solving Eq. 2-27 with the methods and parameters given in Chapter 2-3, we are able to simulate the refractive index profiles and mode profiles of APE MgO:LiNbO₃ waveguides with different PE and annealing time.



Fig. 2-3. Calculated refractive index of APE MgO:LiNbO₃ with 2 hours of PE time, and 0~6 hours of annealing time.



Fig. 2-4. (a) simulated and (b) measured waveguide mode profile of a 6µm wide APE MgO:LiNbO₃ ridge waveguide fabricated with 2 hours PE time and 6 hours of annealing time.



Fig. 2-5 (a) horizontal and (b) vertical cross-section mode profiles of results shown in Fig. 2-4. Solid line is the simulated result, and dotted line is the measured result.

To compare the simulations with experiments, we fabricated a 6 μ m wide APE MgO:LiNbO₃ ridge waveguide with 2 hours of PE time at 200 °C and 6 hours of annealing time at 350 °C. Fig. 2-3 shows the calculated refractive index profile of the waveguide at 1064nm wavelength. Fig. 2-4 and 2-5 shows the simulated and measured mode profiles at 1064nm. The simulation result matches well with the measurement.

2-5. Summary

In this chapter, we have established a basic simulation method of PPLN devices using finite-difference method on three-wave interaction equations. The refractive index model has also been developed for APE waveguides. Using these calculations, we can roughly yet efficiently estimate the theoretical performance of a PPLN waveguide.

REFERENCES:

- [2-1]. O. Gayer, Z. Sacks, E. Galun, and A. Arie, "Temperature and wavelength dependent refractive index equations for MgO-doped congruent and stoichiometric LiNbO₃", Appl. Phys. B 91, 343 (2008).
- [2-2]. M. L. Bortz and M. M. Fejer, "Annealed proton-exchanged LiNbO₃ waveguides", Opt. Lett. 16, 1844 (1991).

Chapter 3. Characterization of APE MgO:LiNbO₃ Using FTIR Spectroscopy

3-1. Annealed Proton-Exchange in LiNbO₃ and MgO:LiNbO₃

The proton-exchange (PE) process in LiNbO₃ has been well studied in the literature. In 1982, J. L. Jackel, C. E. Rice, and J. J. Veselka found that a light-guiding thin film was formed on the surface of a LiNbO₃ crystal after immersing in melted benzoic acid [3-1], due to ion exchanges between H⁺ and Li⁺. Hydrogen ions are exchanged into the LiNbO₃ crystal, forming an $H_xLi_{1-x}NbO_3$ composite. It was found that the compound possesses higher extraordinary and lower ordinary refractive indexes than the bulk LiNbO₃ crystal, and such changes are related to lattice distortions created by the incorporated hydrogen ions. [3-2]-[3-4]. The hydrogen concentrations and its distribution profiles in the composite can be measured by the secondary ion mass spectrometry (SIMS) [3-5]-[3-7]. In benzoic acid proton-exchange, the PE layer possesses a step-like profile, and its thickness follows the Arrhenius relationship. The layer thickness is proportional to the square root of the PE time [3-8], [3-9]. However, SHG experiments in PE LiNbO₃ waveguides confirmed that the nonlinear coefficient d_{33} dropped to zero, due to large lattice distortions at high hydrogen concentration. [3-10]. Later, researchers found that the nonlinear coefficient can be restored as much as 90% of its bulk value by thermal annealing [3-8] [3-11]. During annealing, the protons in the PE layer diffuse into the substrate and create a gradient profile. Further studies using X-ray diffraction have found seven crystallographic phases in PE and APE LiNbO₃ processed under various fabrication conditions. [3-6] [3-7] [3-12] [3-13]. The formation of such phases is closely linked with hydrogen concentrations and processing temperatures. Post PE annealing decreases the

hydrogen concentration in the crystal, and the crystal will eventually end up in the α phase, which possesses the lowest hydrogen concentration with minimal lattice distortions. The fraction of hydrogen ions is x<0.12 in the α phase H_xLi_{1-x}NbO₃. It was found that APE LiNbO₃ waveguides with low propagation loss and high second-order nonlinear coefficient can be obtained with the α phase crystals [3-8] [3-9] [3-11] [3-13].

Infrared (IR) spectroscopy has also been adopted in analyzing the structural and optical characteristics of APE LiNbO₃ waveguides. After proton-exchange, a strong absorption peak at 3510cm⁻¹ was observed due to OH⁻ stretching vibrations [3-13] [3-14] After annealing, the absorption peak moved to 3480cm⁻¹. It has been confirmed that the absorption peak at 3510cm⁻¹ and 3480cm⁻¹ belongs to the β (0.44<x<0.71) phase and the α phase (*x*<0.12) H_xLi_{1-x}NbO₃ crystals respectively [3-13].

Similarly, APE MgO:LiNbO₃ waveguides can be fabricated and characterized using the same techniques. However, it has been found that APE MgO:LiNbO₃ waveguides possess many unique characteristics. J. L. Jackel [3-15], and M. Digonnet *et al.* [3-16] reported that the hydrogen diffusion coefficient in MgO:LiNbO₃ is smaller than that in LiNbO₃. J. L. Jackel [3-15] and A. Loni *et al.* [3-17] reported that the OH⁻ absorption peak of MgO:LiNbO₃ is different from that in none-doped LiNbO₃ . Yu. N. Korkishko *et al.* [3-18] performed X-ray and IR spectrum studies on MgO:H_xLi_{1-x}NbO₃ and found no clear conclusion of phase transitions. The structural properties of MgO:H_xLi_{1-x}NbO₃ [3-18]. By measuring the refractive index profiles of Y-cut APE MgO:LiNbO₃ waveguides, X. Cao *et al.* found that a step-like index profile still existed after 20 hours of annealing due to lower thermal diffusion coefficients [3-19].

Today, although APE MgO:LiNbO3 waveguides are already commercially available,

the mechanisms behind them is not yet fully understood. Till now, there is no systematic study of PE and annealing in MgO:LiNbO₃ in literature. The differences between none-MgO-doped APE LiNbO₃ and APE MgO:LiNbO₃ need to be studied. Through comparison, we can study the PE characteristics and find optimized fabrication conditions for APE MgO:LiNbO₃ waveguides. More importantly, the recovery of nonlinear optical coefficients in APE MgO:LiNbO₃ need to be investigated to yield satisfactory nonlinear conversion efficiency.

In this chapter, we employed the Fourier transform infrared (FTIR) spectroscopy technique, and investigated the phase transitions in APE MgO:LiNbO₃ during thermal annealing. The results were compared with none-doped APE LiNbO₃ crystals. We also prepared an APE MgO:PPLN waveguide for tests. By measuring its second harmonic generation (SHG) properties, the relationships between phase transitions and recovery of nonlinear coefficients were studied. As a result, we can obtain a general idea of how to optimize the performances of APE MgO:PPLN waveguides in our experiments.

3-2. Sample Preparation and Test Conditions

Commercial z-cut LiNbO₃ and 5 mol% MgO doped LiNbO₃ wafers were used in the experiments. Each wafer was diced into 1cm by 1cm square samples. The samples were processed using procedures described in Chapter 4. The PE temperature is 200°C, and the annealing temperature is 350°C. The PE time for each sample varied from 0 hour to 24 hours. The accumulated annealing time for each sample was up to 24 hours.

The FTIR measurements were carried out using a non-polarized SiC light source. The beam direction was perpendicular to the z-cut crystal surface, or parallel to the crystal's

z-axis. The resolution of the FTIR spectrometer was set to 4cm^{-1} wavenumber, or approximately 3nm at the 3500cm⁻¹ band. Each data point took average of 10 scans.

3-3. Comparison of PE LiNbO₃ and PE MgO:LiNbO₃

The measured FTIR spectra of PE LiNbO₃ and PE MgO:LiNbO₃ after are shown in Fig. 3-1(a) and Fig. 3-2 (a) respectively. In Fig. 3-1 (a), the none-processed LiNbO₃ crystal (0 PE hour) shows a small absorption peak centered at 3479cm⁻¹. Such absorption is caused by minute amount of OH⁻ incorporated during crystal growth. After PE, an absorption peak centered at 3501cm⁻¹ appears, and its absorption strength increases with the PE time. The absorption peak is also attached with a broadband tail extending to lower wavenumbers. The measured results agreed well with literatures, and the 3479cm⁻¹ and 3501cm⁻¹ absorption signatures correspond to the α and β phase H_xLi_{1-x}NbO₃ respectively [3-13] [3-20].

In contrast, in Fig. 3-2 (a), the absorption spectra of none-processed MgO:LiNbO₃ show very distinctive features. The spectrum displays an absorption peak centered at 3533cm⁻¹, while carries a tiny absorption peak at 3479cm⁻¹. However, similar to LiNbO₃, an absorption peak located at 3501cm⁻¹ is also present after PE. The results agreed with previous findings on OH⁻ absorption band locations in PE MgO:LiNbO₃ [3-15] [3-17] [3-19]. However, unlike what reported in literature, the absorption peak does not shift from 3533cm⁻¹ to 3501cm⁻¹; instead, the 3533cm⁻¹ peak is a new addition to the spectrum, which is clearly visible when comparing the curves of none-processed MgO:LiNbO₃ and PE MgO:LiNbO₃ in Fig. 3-2 (a). Because the 3501cm⁻¹ absorption is relatively strong and broadband, the three absorption peaks have merged together.

The obtained spectrum information suggests that the PE process introduces the same structural changes to both none-doped LiNbO₃ and MgO:LiNbO₃. It is also very likely that the MgO: $H_xLi_{1-x}NbO_3$ compound possesses at least one additional phase, which does not exist in the none-doped crystal.

The proton diffusion behaviors in none-MgO-doped APE LiNbO₃ have been well studied in literature. The PE layer thickness can be calculated as $d = 2\sqrt{D(T) \cdot t}$, where D(T) is the temperature-dependent diffusion coefficient, and *t* is the PE time [3-21]. Since light transmission in PE LiNbO₃ follows the Beer-Lambert law, we can calculate the spectrum transmission at 3501cm⁻¹ as:

$$T = e^{-2\sigma\sqrt{D}\cdot t} \tag{3-1}$$

where σ is the absorption coefficient of the β phase H_xLi_{1-x}NbO₃. Because PE MgO:LiNbO₃ has demonstrated an identical absorption signature as the β phase H_xLi_{1-x}NbO₃ did at the 3501cm⁻¹ band, it is safe to predict that their crystal phases and hydrogen concentrations are very similar. As a result, it is reasonable to assume:

$$\sigma_{PE Mg0:LiNb0_3} = \sigma_{PE LiNb0_3} \tag{3-2}$$

Using the above assumption, we are able to perform curve fitting using the measured transmission data at 3501 cm⁻¹, and the results are shown in Fig. 3-1 (b) and Fog. 3-2 (b). We can calculate the ration of diffusion coefficient in MgO:LiNbO₃ and none-MgO-doped LiNbO₃ is approximately $D_{MgO:LiNbO_3}/D_{LiNbO_3} = 0.76$. This agrees with earlier literatures that found proton-exchange in MgO:LiNbO₃ is slower than that in none-MgO-doped LiNbO₃ [3-15] [3-17]. For none-MgO-doped LiNbO₃, we can find the proton diffusion coefficient at 200°C is $D(200°C)=0.081\mu m^2/h$ [3-16]. Hence the proton diffusion coefficient for MgO:LiNbO₃ at 200 °C is $D(200°C)=0.062\mu m^2/h$.



Fig. 3-1. (a). Absorption spectra of none-doped PE LiNbO₃. (b). Measured light transmittance at 3501cm⁻¹ (solid), and exponential numerical fitting (dashed) in PE LiNbO₃



Fig. 3-2. (a). Absorption spectra of PE MgO:LiNbO₃. (b). Measured light transmittance at 3501cm⁻¹ (solid), and exponential numerical fitting (dashed) in PE MgO:LiNbO₃.

3-4. Comparison of APE LiNbO₃ and APE MgO:LiNbO₃

After PE, the samples were annealed up to 24 hours. Fig. 3-3 shows the evolution of absorption spectrum in none-MgO-doped LiNbO₃ and MgO:LiNbO₃ during annealing. The samples both have 4 hours of PE time. Fig. 3-3 (a) shows that after annealing, the 3501cm⁻¹ peak decreased while the 3479cm⁻¹ peak increased. This corresponds to the phase transition from β to α in APE LiNbO₃ [3-20]. Similarly, in Fig. 3-3 (b), a phase transition from 3501cm⁻¹ to 3479cm⁻¹ was also detected in APE MgO:LiNbO₃. However, the absorption peak at 3533cm⁻¹ remains and becomes stronger with increased annealing time. To avoid confusion with the none-MgO-dope APE LiNbO₃ system, we name the phases in APE MgO:LiNbO₃ as phase α , b and c, corresponding to absorption bands at 3479cm⁻¹, 3501cm⁻¹ and 3533cm⁻¹.

Since the absorption peaks are partially overlapped with each other, to better evaluate the evolution of different phases during annealing, curve fitting is performed to de-convolute the absorption spectra following the Beer-Lambert law. In Fig. 3-1 (a), the non-processed (0 hour PE time) LiNbO₃ has provided the absorption curve for phase α or phase α crystal. By subtracting the absorption curve of phase α crystal from the absorption curve with 2 hours PE time, we get the absorption curve of phase β or phase bcrystal. In Fig. 3-2 (a), the absorption bands of phase α and phase c crystals in the none-processed MgO:LiNbO₃ are not overlapped. After separation, we get the absorption curve of phase c crystal.

Under weak absorption conditions, the area covered under each absorption curve should equal to each other, for each equal amount of OH^- groups acting as absorbers. Following the above assumptions, Fig. 3-4 shows the normalized absorption curves of phase *a*, *b*, and *c* crystals with the same amount of active OH^- groups. Using these

absorption curves as templates, we can perform curve fitting following the Beer-Lambert law. An equivalent layer thickness d_a , d_b and d_c is assigned to phase a, b and c crystals respectively, so that the transmission can be written as $T = e^{-\sigma(d_a+d_b+d_c)}$. Fig. 3-5 shows the curve fitting procedures for a sample with 4 hours of PE time and 8 hours of annealing time. By varying the amount of phase a, b, and c crystals, we can obtain a close fit to each measured spectrum. After curve fitting, we can obtain the equivalent amount of phase α and β crystals in APE LiNbO₃, and phase a, b and c crystals in APE MgO:LiNbO₃ with different PE and annealing time. The results are shown in Fig. 3-6 to Fig. 3-10. The normalization unity is set at 24 hours PE in LiNbO₃, where maximum amount of OH groups are produced.

In APE LiNbO₃, the phase transition stopped after phase β crystals were depleted; and the amount of phase α crystals became stable. Samples with 4 and 8 hours of PE time reached depletion within 24 hour of annealing time, while samples with longer PE time needed longer annealing time.

Similar depletion of phase *b* crystals was observed in APE MgO:LiNbO₃. However, in APE MgO:LiNbO₃, the amount of phase *a* crystals slightly decreased after the depletion of phase *b* crystals, which is very different from APE LiNbO₃. Throughout the annealing process, the amount of phase *c* crystals kept a slow but steady growth. Combining with the comparison results of the non-processed LiNbO₃ and MgO:LiNbO₃ crystal in Fig. 3-1 (a) and Fig. 3-2 (a), we can conclude that in APE MgO:LiNbO₃, the phase *c* crystal has the lowest hydrogen concentration. Although the phase *a* crystal in APE MgO:LiNbO₃ possesses the same spectrum signature as the phase *a* crystal in LiNbO₃, it is not the most stable phase in APE MgO:LiNbO₃. It will eventually transfer into phase *c*, given sufficient time and temperature. However, from Fig. 3-8 and Fig. 3-10,



we have noticed that such transformation is extremely slow.

Fig. 3-3. Absorption spectra of (a) APE LiNbO₃ and (b) APE MgO:LiNbO₃ with 4 hours of proton-exchange time and 0~24 hours of annealing time.



Fig. 3-4. Normalized absorption spectrum of phase *a*, *b*, and *c* crystals in APE MgO:LiNbO₃.



Fig. 3-5. Curve fitting result of an APE MgO:LiNbO₃ sample with 4 hours of PE time and 8 hours of annealing. Individual absorption curves from different amount of phase *a*, *b*, *c* crystals are shown.



Fig. 3-6. Amount of phase α crystals in APE LiNbO₃ during annealing. The PE time on each sample is 0~24 hours



Fig. 3-7. Amount of phase β crystals in APE LiNbO₃ during annealing. The PE time on each sample is 0~24 hours



Fig. 3-8. Amount of phase *a* crystals in APE LiNbO₃ during annealing. The PE time on each sample is 0~24 hours



Fig. 3-9. Amount of phase *b* crystals in APE LiNbO₃ during annealing. The PE time on each sample is $0\sim24$ hours



Fig. 3-10. Amount of phase *c* crystals in APE LiNbO₃ during annealing. The PE time on each sample is 0~24 hours



Fig. 3-11. X-ray diffraction results of APE MgO:LiNbO₃ crystals

To confirm the disappearance of phase *b* crystals in well-annealed MgO:LiNbO₃ from a different perspective, we performed an x-ray diffraction measurement on several samples. Figure 3-11 shows the x-ray rocking curve of MgO:LiNbO₃ without PE, after 4 hours of PE, and after 24 hours of annealing. The measurement was performed using a Cu K-alpha source at 0.154nm wavelength, and the diffraction plane was set at the (0, 0, 12) with a Bragg angle of 41.8175°.

The untreated MgO:LiNbO₃ crystal shows a strong diffraction peak at the Bragg angle (position 0). After PE, the phase β or phase *b* crystals introduced a large lattice distortion to the crystal, and produced additional diffraction peaks at around -700 arcsec position. The result agreed with previous reported research data in PE LiNbO₃ [3-18]. After 24 hours of annealing, the diffraction peaks at around -700 arcsec disappeared completely, which suggested a complete depletion of phase β or phase *b* crystals. Meanwhile, we noticed the diffraction peak at position 0 was slightly broader after APE, which was caused by the slight lattice distortion introduced by the phase *a* and phase *c* crystals. This also agreed with previous studies on phase *a* crystals in APE LiNbO₃ [3-18].

3-5. Recovery of Nonlinear Coefficient in APE MgO:LiNbO₃

Similar to none-doped APE LiNbO₃, we expect a recovery of the d₃₃ nonlinear coefficient during the thermal annealing of APE MgO:LiNbO₃. It has been widely recognized that the phase α APE LiNbO₃ is suitable for low-loss, high efficiency nonlinear optical waveguides [3-9]. In APE MgO:LiNbO₃, the existence of phase *c* crystals has added complexity to the waveguide's structures, and made it different from

none-MgO-doped APE LiNbO₃. To better understand the behaviors of phase *a*, *b*, and *c* crystals, their nonlinear optical properties must be examined. In order to optimize the performance of APE MgO:LiNbO₃ waveguides, it is necessary to find out the annealing time needed to recover the d_{33} nonlinear coefficient.

We prepared a z-cut APE MgO:PPLN slab waveguide using the same procedures described in Chapter 4. Because of the availability of MgO:PPLN crystals and cost concerns, only one piece of sample is fabricated. The PE time was set at 4 hours, and the annealing time was from 0 to 24 hours.



Fig. 3-12. SHG spectra of the MgO:PPLN slab waveguide with 4 hours of proton exchange and 0~24 hours of annealing time.



Fig. 3-13. (a). Output power of the APE MgO:PPLN slab waveguide. (b). SHG signal strength and normalized conversion efficiency of the waveguide.

By measuring the SHG signals of the APE MgO:PPLN waveguide, we can evaluate the recovery of the d_{33} second-order nonlinear coefficient during annealing. We use a 1.5µm broadband light source as an input. Light was coupled into and out of the waveguide through single mode fiber. A spectrum analyzer and power meter were used to read the output signals. Fig. 3-12 shows the obtained SHG spectrum of the slab waveguide through 0~24 hours of annealing.

We noticed that the SHG signal increases from 2 hours to 10 hours, and then keeps steady to 24 hours of annealing time. The spectrum positions of the SHG signals are also changing significantly below 10 hours of annealing, due to the evolution of refractive index and the modal dispersion relationships, which changes the SHG quasi-phase matching condition.

Fig. 3-13 (a) shows the measured output power of the waveguide. The tests were conducted under low-power conditions (<1mW). The output is mainly fundamental light. Although we have found a steady growth of output power of fundamental light after 0, 0.5, and 1 hour of annealing time, no SHG signal was obtained. The waveguide output power, which reflects the waveguide insertion loss, becomes steady after 12 hours of annealing. We did not separate the waveguide propagation loss and the coupling loss in our experiment, considering the overall performance of the waveguide with fiber systems.

Fig. 3-13 (b) shows the measured SHG signal strengths from Fig. 3-12, and the normalized SHG efficiencies calculated using:

$$\eta = P_{2\omega} / P_{\omega}^2 \ (\%/W) \tag{3-3}$$

where P_{ω} is the power of the fundamental light, and $P_{2\omega}$ is the power of the SHG light. In Fig. 3-13 (b), the large value anomaly at 2 hours of annealing is likely caused by large waveguide propagation loss. Since P_{ω} and $P_{2\omega}$ are both measured at the output end of the waveguide, the SHG efficiency was magnified by $\eta' = (\alpha P_{2\omega})/(\alpha P_{\omega})^2$, where α is the waveguide loss coefficient, and $\alpha < 1$.

From Fig. 3-12 and Fig. 3-13, we noticed that after 10 hours of annealing, the waveguide reaches steady SHG spectrum position, output power, and conversion efficiency. Comparing the result with those from Fig. 3-8 to 3-10, we discovered that the stabilization of the SHG signal for the waveguide with 4 hours of PE time occurred where the phase transition from phase *b* to phase *a* was completed, and the amount of phase *a* crystals stopped increasing. The above findings indicate that the phase *b* crystal in PE and APE MgO:LiNbO₃ possesses a near-zero d_{33} value, and the phase *a* crystal possesses a recovered non-zero d_{33} value. The behaviors of phase *a* and *b* crystals in APE MgO:LiNbO₃ are very similar to that of phase *a* and *β* crystals in APE LiNbO₃. Following the relationship between hydrogen concentrations and nonlinear optical coefficients in APE LiNbO₃ [3-8] [3-11], the d_{33} value of the phase *c* crystal is likely to be non-zero as well. Phase *a* and phase *c* crystals co-exist in well-annealed APE MgO:LiNbO₃ waveguides.

From the FTIR results shown in Fig. 3-9, we can estimate the annealing time needed for APE MgO:LiNbO₃ samples with different PE times to reach stabilized performance. For each curve, we can find the time point where $\partial y/\partial x=0$, which indicates the depletion of phase *b* crystals. According to the analysis of the MgO:PPLN sample with 4 hours of PE time, we can estimate the nonlinear optical performance of APE MgO:PPLN crystals with other PE times. Stable performances of SHG phase matching condition, output power, and conversion efficiency can be obtained using the annealing time indicated in Fig. 3-14. In practical situations, SHG devices with optimized performances can be

engineered following this PE time and annealing time relationship, with slight tailoring according to the operating wavelengths and other specifications of the application.



Fig. 3-14. Relationship between the PE time and the annealing time for APE MgO:LiNbO₃ waveguide to reach phase *b* depletion and stable performance.

3-6. Summary

In this chapter, we have characterized the absorption spectrum of APE LiNbO₃ and APE MgO:LiNbO₃ crystals. Although the APE process in MgO:LiNbO₃ is very similar to that in LiNbO₃, there are a few distinctive differences. The evolutions of three absorption peaks at 3501cm⁻¹, 3479cm⁻¹ and 3533cm⁻¹ during thermal annealing indicate that there are at least three distinctive phases in the MgO:H_xLi_{1-x}NbO₃ composite. The hydrogen concentration in APE MgO:LiNbO₃ is phase *b*>phase *a*>phase *c*. We monitored the SHG

performance of an APE MgO:LiNbO₃ waveguide during annealing. Through the SHG tests, we found that the d_{33} value in phase *b* is near-zero, while it is non-zero in phase *a* and phase *c* crystals. After a certain amount of annealing time, the APE MgO:LiNbO₃ waveguide can reach optimized performance, where the waveguides are mainly consisted of phase *a* and *c* crystals. Combining the measurement of the FTIR spectra and SHG signals, we have found a relationship between the PE time and the annealing time for optimizing the performance of APE MgO:LiNbO₃ waveguides.

REFERENCES:

- [3-1]. J. L. Jackel, C. E. Rice, and J. J. Veselka, "Proton-exchange for high-index waveguides in LiNbO₃," Appl. Phys. Lett. 41, 607 (1982).
- [3-2]. K. Ito and K. Kawamoto, "Dependence of lattice constant deviation and refractive index on proton concentration in proton-exchanged optical waveguides on a single crystal of LiNbO₃", Jpn. J. Appl. Phys. 31, 3882 (1992).
- [3-3]. M. M. Howerton, W. K. Burns, P. R. Skeath, and A. S. Greenblatt, "Dependence of refractive index on hydrogen concentration in proton exchanged LiNbO₃", IEEE J. Quant. Electron. 27, 593 (1991).
- [3-4]. J. M. Zavada, H. C. Casey Jr., R. J. States, S. W. Novak, and A. Loni, "Correlation of substitutional hydrogen to refractive index profiles in annealed proton-exchanged Z- and X-cut LiNbO₃", J. Appl. Phys. 77, 2697 (1995).
- [3-5]. C. Canali, A. Carnera, G. Della Mea, P. Mazzoldi, S. M. Al Shukri, A. C. G. Nutt, and R. M. De La Rue, "Structural characterization of proton exchanged LiNbO₃

optical waveguides", J. Appl. Phys. 59, 2643 (1986).

- [3-6]. R. G. Wilson, S. W. Novak, J. M. Zavada, A. Loni and R. M. De La Rue, "Secondary ion mass spectrometry depth profiling of proton-exchanged LiNbO₃ waveguides", J. Appl. Phys. 66, 6055 (1989).
- [3-7]. A. Loni, R.M. de la Rue, J.M. Zavada, R.G. Wilson, and S.W. Novak, "Direct comparison of hydrogen depth distributions and refractive index profiles in annealed proton-exchanged z-cut LiNbO₃ waveguides", Electron. Lett. 27, 1245 (1991).
- [3-8]. X. Cao, R. Srivastava, R. V. Ramaswamy, and J. Natour, "Recovery of second-order optical nonlinearity in annealed proton-exchanged LiNbO₃", IEEE Photon. Technol. Lett. 3, 25 (1991).
- [3-9]. K. R. Parameswaran, R. K. Route, J. R. Kurz, R. V. Roussev, M. M. Fejer, and M. Fujimura, "Highly efficient second-harmonic generation in buried waveguides formed by annealed and reverse proton exchange in periodically poled lithium niobate", Opt. Lett. 27, 179 (2002).
- [3-10]. M. L. Bortz and M. M. Fejer, "Measurement of the second-order nonlinear susceptibility of proton-exchanged LiNbO₃", Opt. Lett. 17, 704(1992).
- [3-11]. M. L. Bortz, L. A. Eyres, and M. M. Fejer, "Depth profiling of the d₃₃ nonlinear coefficient in annealed proton exchanged LiNbO₃ waveguides", Appl. Phys. Lett. 62, 2012 (1993).
- [3-12]. C. Canali, A. Carnera, G. Della Mea, P. Mazzoldi, S. M. Al Shukri, A. C. G. Nutt, and R. M. De La Rue, "Structural characterization of proton exchanged LiNbO₃

optical waveguides", J. Appl. Phys. 59, 2643 (1986).

- [3-13]. C. E. Rice, "The structure and properties of Li_{1-x}H_xNbO₃", J. Solid State Chem. 64, 188 (1986).
- [3-14]. A. Loni, G. Hay, R. M. De La Rue, and J. M. Winfield, "Proton-exchanged LiNbO₃ waveguides: the effects of post-exchange annealing and buffered melts as determined by infrared spectroscopy, optical waveguide measurements, and hydrogen isotopic exchange reactions", IEEE J. Lightwave Technol. 7, 911 (1989).
- [3-15]. J. L. Jackel, "Proton exchange in MgO-doped LiNbO₃", Electron. Lett. 21, 509 (1985).
- [3-16]. M. Digonnet, M. Fejer, and R. Byer, "Characterization of proton-exchanged waveguides in MgO:LiNbO₃", Opt. Lett. 10, 235 (1985).
- [3-17]. A. Loni, R. W. Keys, and R. M. De La Rue, "Characterization of waveguides formed by proton-exchange in MgO-doped and Nd:MgO-doped LiNbO₃:A comparison with congruent material", J. Appl. Phys. 67, 3964 (1990).
- [3-18]. Yu. N. Korkishko, V. A. Fedorov, S. M. Kostritskii, E. I. Maslennikov, M. V. Frolova, A. N. Alkaev, C. Sada, N. Argiolas and M. Bazzan, "Proton-exchanged waveguides in MgO-doped LiNbO₃: optical and structural properties", J. Appl. Phys. 94, 1163 (2003).
- [3-19]. X. Cao, Z. Wang, S. He, L. Zhan, and Y. Xia, "Optical and structural characterization of annealed proton exchange waveguides in Y-cut MgO:LiNbO₃", Opt. Mater. 27, 1596 (2005).
- [3-20]. Yu. N. Korkishko and V. A. Fedorov, "Relationship between refractive indices and

hydrogen concentration in proton exchanged waveguides", J. Appl. Phys. 82, 1010 (1997).

[3-21]. M. L. Bortz and M. M. Fejer, "Annealed proton-exchanged LiNbO₃ waveguides", Opt. Lett. 16, 1844 (1991).

Chapter 4. Fabrication Techniques of APE MgO:PPLN Ridge Waveguide

4-1. Fabrication Procedures of APE MgO:LiNbO₃

The proton-exchange (PE) process was carried out in melted benzoic acid (melting point 122.4 °C) at 200 °C. Z-cut 5mol% MgO doped LiNbO₃ crystals were used in the experiment. The crystals and benzoic acid were put into a crucible, and then placed into a temperature-controlled furnace. The crucible was covered but not sealed. After a certain amount of PE time, the crucible was removed from the furnace and cooled to room temperature. The crystals were extracted and cleaned with acetone and ethanol. The annealing process was performed in a temperature-controlled tube furnace purged with oxygen. The annealing temperature was set at 350 °C. The PE and annealing temperatures were optimized conditions to produce low-loss APE LiNbO₃ waveguides with recovered second-order nonlinear coefficient [4-1]-[4-4]. Fig. 4-1 shows the fabrication procedures.



Fig. 4-1. Fabrication procedures of APE MgO:PPLN crystals. The PE process is carried out at 200 ℃, the annealing process is performed at 350 ℃.

4-2. Ridge Structure Formation

After the APE process, ridge structures were formed by precise diamond blade dicing. The dicing operations were performed on an automatic dicing saw. The dicing saw has a step accuracy of 2µm. A dicing blade with diamond particles of 2µm average diameter was used to ensure optical grade finish. The ridge width can be flexibly chosen by each application. The ridge height was set to 30µm to avoid the rounding corners on the blade's tip and ensure near 90° sidewall finish. The waveguide's facets were processed using a similar precise dicing method. The dicing depth was set at 70µm for direct coupling of optical fibers with 125µm diameter. Fig. 4-2 shows the dicing results of several 6µm wide 30µm deep ridge waveguides. Later in our experiment, we found that the 30µm ridge height was not necessary for maintaining a perpendicular sidewall, and reduced it to 20µm.

In order to quantitatively assess the waveguide quality, we performed a series of measurements. Fig. 4-3 shows a waveguide sample measured with scanning electron microscope (SEM) and atomic force microscope (AFM). The SEM measurement shows that the waveguide sidewall has a 93 $^{\circ}$ angle. The AFM measurement shows that the surface roughness of the diamond blade diced facets has a root-mean-square (RMS) value of 3.72nm. We measured the waveguide propagation loss by comparing the insertion losses of waveguides with different lengths. An Yb-doped broadband light source and an Er-doped broadband light source were used in the tests to assess the waveguide losses at 1.0 μ m-band and 1.5 μ m-band wavelengths respectively. The measured waveguide propagation loss was about 0.2dB/cm for both wavelength bands.



Fig. 4-2. MgO:PPLN ridge waveguides processed by precise diamond blade dicing. The ridges are about 6μm wide and 30μm high. The waveguide facets are 70μm high.



Fig. 4-3. (a) SEM images of a MgO:LiNbO₃ ridge waveguide. The red square in the lower left part indicates the area of the AFM test. (b) and (c) are the AFM test result. The measured surface roughness RMS value is 3.72 nm.

4-3. Repeatability of the Dicing Process

The precise dicing method uses diamond particles in the dicing blade to grind and polish the crystal. As a result, the blade will wear out after a certain time of use. Regular maintenance and inspections of the dicing blade is crucial for stable output. Fig.4-4 shows several situations when the blade is mildly or heavily worn. The waveguides show asymmetric profiles or irregular stripes on the sidewall.



Fig. 4-4. SEM image of MgO:PPLN ridge waveguides processed under improper conditions. The ridges show deteriorated results with asymmetric profiles in (a) (b), and irregular stripes in (c) (d).
When properly maintained, the dicing saw can yield very stable output. Fig. 4-5 shows a MgO:PPLN crystal that contains 19 ridge waveguides. The chip length is 4cm. The waveguides show no measureable profile variations across the chip.



Fig. 4-5. Photos of a 4cm long MgO:PPLN crystal that contains 19 ridge waveguides.

4-4. Summary

In this chapter, we have demonstrated successful fabrication techniques of APE MgO:LiNbO₃ ridge waveguides. The ridge waveguides have surface roughness less than

4nm, and side walls near 90°. Good uniformity has been achieved over 4cm length. The waveguides have low propagation loss of about 0.2dB/cm. The properties of the waveguides can be easily tuned by changing the APE or precise machining parameters. Comparing with other types of waveguides, the fabrication techniques combine together flexibility, repeatability, high efficiency, and low cost.

REFERENCES:

- [4-1]. M. L. Bortz, L. A. Eyres, and M. M. Fejer, "Depth profiling of the d₃₃ nonlinear coefficient in annealed proton exchanged LiNbO₃ waveguides", Appl. Phys. Lett. 62, 2012 (1993).
- [4-2]. Yu. N. Korkishko and V. A. Fedorov, "Relationship between refractive indices and hydrogen concentration in proton exchanged waveguides", J. Appl. Phys. 82, 1010 (1997).
- [4-3]. C. E. Rice, "The structure and properties of Li_{1-x}H_xNbO₃", J. Solid State Chem. 64, 188 (1986).
- [4-4]. K. R. Parameswaran, R. K. Route, J. R. Kurz, R. V. Roussev, M. M. Fejer, and M. Fujimura, "Highly efficient second-harmonic generation in buried waveguides formed by annealed and reverse proton exchange in periodically poled lithium niobate", Opt. Lett. 27, 179 (2002).

Chapter 5. 1064nm to 532nm Second Harmonic Generation

5-1. Application Background

Compact high power green lasers are highly demanded in various applications such as laser projection display and biomedical instrumentation. Although semiconductor green laser diodes have been reported in recent years [5-1]-[5-3], to date, wavelength conversion via nonlinear optical crystals remain the most widely used method to obtain high power green light. The MgO:PPLN crystal has been proved as an efficient wavelength conversion material in milliwatt to watt-level power range.

Single-pass green light SHG using MgO:PPLN waveguides has attracted much attention in recent years. Several companies and research institutes have developed highly efficient green laser modules based on waveguide designs. K. Sakai et al. from Mitsubishi Electric Corporation reported 1.6W green light generation from a 150µm wide MgO:PPLN slab waveguide with 60% optical conversion efficiency [5-4] [5-5]. H. K. Nguyen et al. from Corning Incorporated reported an 304mW green light generation from an MgO:PPLN ridge waveguide with an optical conversion efficiency of 72.9% [5-6]. However, the above-mentioned works are based on wafer-bonded waveguide, and the fabrication costs are high.

In order to develop a green laser with compact size, high output power, high efficiency, and low fabrication cost, we designed the APE MgO:PPLN ridge waveguide for 532nm light generation. We use a low-cost linearly polarized fiber laser as the source of 1064nm light. The 1064nm light is converted to 532 nm light through QPM-SHG in the waveguide.

5-2. Waveguide Design and Fabrication

Since the waveguide is designed to be used with a fiber laser, the fiber laser was characterized first. Fig. 5-1 shows the output spectrum of the fiber laser. The measured full width half maximum (FWHM) bandwidth of the laser is 60pm, and the central wavelength is 1063.38nm.



Fig. 5-1. Spectrum of the 1064nm fiber laser used for green light SHG. The laser has a FWHM bandwidth of 60pm, and central wavelength of 1063.38nm.

The laser was built with polarization maintaining (PM) fiber with a mode field diameter (MFD) of 6.6 μ m. In order to maximize the coupling efficiency from the fiber laser to the waveguide, the waveguide width was designed to be 8 μ m. The obtainable waveguide dimension was limited the dicing saw's 2 μ m step accuracy, which means the available numbers were 6 μ m, 8 μ m, or 10 μ m by setting. Because the APE waveguide only supports TM modes, the fiber laser's output polarization is aligned to vertical

direction by physically rotating the PM fiber.

The APE conditions are set to 2 hours PE at 200 °C, and 6 hours annealing at 350 °C to obtain an optimized performance with recovered d_{33} nonlinear coefficient. After fabrication, the obtained waveguide length is 14mm. Fig. 5-2 shows calculated refractive index profiles of the waveguide at 1064nm and 532nm.

Since the waveguide facets are uncoated, the Fresnel reflections are as high as 13%. As a result, parallel facets will cause significant Fabry-Perot (F-P) effect, which in turn cause wavelength and temperature dependent power fluctuations in the waveguide. We calculated the F-P interference patterns inside the waveguide, and found the F-P frequency comb spacing is 13.4pm at 1064nm and 3.3pm at 532nm. Since the line width of our 1064nm laser is much broader, we found the F-P effect cannot cause noticeable transmission fluctuation.



Fig. 5-2. Calculated refractive index profile for APE MgO:LiNbO3 waveguide at (a) 1064nm, and (b). 532nm. The waveguide is processed with 2 hours of PE and 6 hours of annealing time.

5-3. Green Light Generation using APE MgO:PPLN Ridge Waveguide

The experiment setups for testing the waveguide performances are shown in Fig. 5-3. The waveguide chip was temperature-controlled by a Peltier thermal electric controller with 0.1°C accuracy. To reduce the thermal resistance, the waveguide chip was flip-mounted on a silicon substrate with a thin layer of optical adhesives. We have found in previous experiment that during the SHG experiment, there was heat generation from the waveguide, especially under high pump power. Improving heat dissipation is crucial to obtain high output power. The output light was collected by an objective lens, and measured by an optical power meter. A 532nm optical filter was used to eliminate the residue pump light when measuring the green light power.

The measured maximum green light output received at the power meter was 398mW, when the total light output power was 573mW. The uncoated waveguide facet introduced about 14% Fresnel losses to 532nm light. The objective lens and the filter used in the experiment also added a small amount of losses. After calculation, we estimate the green light power generated by the waveguide before exiting the output facet was 466mW, which corresponds to an optical-to-optical conversion efficiency of 69.7%. The power and conversion efficiency curves of the generated green light are shown in Fig. 5-4.



Temperature controlled stage

Fig. 5-3. Experiment setups for 532nm green light SHG using APE MgO:PPLN ridge waveguide. The waveguide chip is flip-mounted to reduce thermal resistance.



Fig. 5-4. Green light power generated by the APE MgO:LiNbO₃ waveguide. The data is calculated with measurement results from the optical power meter and the transmission data of each component after the waveguide.



Fig. 5-5. Measured QPM temperature tuning curves under (a) 100mW, (b) 200mW and (c) 300 mW-level light power.

Although the curve of green light output power shows a tendency for further increasing under higher input power, we were not able to obtain higher output power in experiment. In the contrary, the output power and efficiency started a sharp drop with increased pump. We also noticed that the waveguide's propagation loss increases under higher output power. The Peltier temperature controller also showed higher current, which indicates more heat generation from the waveguide under higher output power.

The waveguide's temperature tuning curves were measured under different output power, which is shown in Fig. 5-5. The curves were obtained by slowly decreasing the waveguide temperature. Under higher pump power, the temperature tuning curve shifted towards lower temperature, and became less symmetric. The waveguide's total output power decreased as the temperature moved towards the phase matching temperature. The temperature controller also registered increased heat generation from the waveguide at phase matching temperatures. Such effects were noticeable with over 100mW light power, and became significant as the light power increased. The results are clear indications of the green induced infrared absorption (GRIIRA) effect, which introduces additional absorption loss to the IR light and subsequent temperature rise due to absorption centers activated by the green light.

5-4. Green Induced Infrared Absorption in MgO:LiNbO₃

Under high light intensity, LiNbO₃ demonstrates considerable amount of photo-induced nonlinear effects such as photorefractive damage (PRD), green induced infrared absorption (GRIIRA), or blue light induced infrared absorption (BLIIRA) [5-7] [5-8]. Such effects cause light absorptions and self-modulated refractive index changes

along the beam propagation direction, which deteriorate the crystal's high power performance. Although doping LiNbO₃ with over 5mol% MgO can greatly reduce the photo-induced effects [5-7]-[5-10], the remaining amount of effects are still strong enough and cannot be ignored in high power lasers.

The origin of photo induced effects in LiNbO₃ have been attributed to certain crystallographic defects, such as $Fe^{2+/3+}$ impurities incorporated during crystal growth [5-11]-[5-13] and Nb_{Li}^{4+/5+} anti-sites generated as intrinsic defects [5-8][5-9]. However, the physics behind such phenomenon have not yet been fully understood. Recent studies have revealed some new contributing mechanisms involved in the process [5-14]- [5-17], especially under MW/cm² to GW/cm² light power densities.

In MgO:PPLN waveguides, the light power density is typically on the order of MW/cm² with several hundred milliwatt input power. M. Iwai *et al.* [5-18] and K. Sakai *et al.* [5-5] have reported power and efficiency saturation effects in crystal-bonded MgO:LiNbO₃ waveguide SHG lasers at 473nm and 532nm respectively. These effects have been attributed to temperature none-uniformity caused by photo-induced light absorptions. In our experiments on APE MgO:PPLN waveguide green lasers, similar problems have been found with the same behavior under increased pump power.

To date, although APE MgO:PPLN waveguides have been widely adopted in nonlinear optical devices, systematic studies on the light induced absorption effects in APE MgO:LiNbO₃ has not been reported yet. In the following chapters, we investigate and compare the GRIIRA characteristics of MgO:LiNbO₃ crystals with APE MgO:LiNbO₃ crystals. By measuring the IR absorption coefficients under the illumination of green light, we can obtain the absorption coefficients of different waveguides under hundreds of kW/cm² to several MW/cm² light power densities. By

- 64 -

quantitatively studying the GRIIRA characteristics, we are able to evaluate heat generations caused by the absorption, and thermal-induced refractive index none-uniformities inside the APE MgO:LiNbO₃ waveguide.

5-5. Measurement of GRIIRA Coefficient in APE MgO:PPLN

To study the GRIIRA effect in APE MgO:PPLN, we first compared the induced absorption coefficient in MgO:LiNbO₃ and APE MgO:LiNbO₃. MgO:LiNbO₃ waveguides with and without APE were prepared. None-APE waveguides were prepared by wafer bonding and precise machining techniques. First, an MgO:LiNbO₃ wafer was bonded to a LiTaO₃ crystal with an optical epoxy. Then the wafer was thinned and polished to about 4µm thick. Finally, ridge structures were fabricated by precise diamond blade dicing. APE waveguides were prepared with 2 hours PE in melted benzoic acid at 200 °C, and 6 hours annealing at 350 °C in oxygen. The length of each waveguide was set to 2cm. Fig. 5-6 shows the SEM photos of the waveguides.



Fig. 5-6. SEM photo of (a). wafer bonded MgO:LiNbO₃ ridge waveguide, and (b). APE MgO:LiNbO₃ ridge waveguide, prepared for the GRIIRA measurements.

The experiment setup for the GRIIRA tests is shown in Fig. 5-7. We used the 1064nm linearly polarized fiber laser described in chapter 5-2 as the IR source, and a diode pumped solid-state 532nm laser as the green light source. The FWHM line width of the IR and green laser is 60pm and 100pm respectively, which was much broader than the F-P fringes introduced by the waveguide facets. As a result, no F-P introduced power fluctuation was observed. The IR laser operated at continuous-wave (CW) output. The 532nm laser was internally modulated by a square-wave pulse trigger set at 1Hz. Since the APE waveguides only support extraordinary polarization, both IR and green light beams were rotated to TM polarization. Each beam was collimated individually, and then combined with a dichroic mirror. The beams were focused by an objective lens and coupled into the ridge waveguide. The output light was collected using another objective lens. The output IR and green light were split using a prism, and the IR and green light power were measured individually.



Fig. 5-7. Experiment setup for the GRIIRA coefficient measurements.

To calculate the light power inside the waveguide, we measured the transmittance of each component after the waveguide at 1064nm and 532nm. The Fresnel reflection of the uncoated waveguide output facet was also considered in the calculation. We took the average power density approach. The mode diameter of the waveguide at each wavelength is measured at $1/e^2$ of the peak power. The effective mode area at 1064nm and 532nm were measured as $18.56\mu m^2$ and $16.33\mu m^2$ for the APE waveguide, and $17.55\mu m^2$ and $15.78\mu m^2$ for the non-APE waveguide. The output mode profiles are shown in Fig. 5-8.

Fig. 5-9 shows a typical IR transmission curve observed in the experiment. The onset of green light pulses triggers a sharp decrease of transmitted IR light by a few percent. The obtained curve is consistent with previous researches on bulk LiNbO₃ [5-8] [5-19]. The turn-on and turn-off time of the absorption is almost instantaneous, which matches with previously reported excitation and relaxation characteristics of the light induced absorption centers. A. Carson *et al.* [5-20] reported the turn-on time of light-induced absorption is several nanoseconds, while the turn-off time is less than 1 millisecond. D. Berben *et al.* [5-21] also reported the relaxation time of photo-induced absorption in LiNbO₃ is in an order of milliseconds. The GRIIRA absorption coefficient was calculated using the average power change as shown in Fig. 5-9 in each measurement, and by applying the Beer-Lambert law.



Fig. 5-8. Measured waveguide output profiles of (a) (b) wafer-bonded MgO:LiNbO₃ waveguide at 1064nm and 532nm, and (c) (d) APE MgO:LiNbO₃ waveguide at 1064nm and 532nm.



Fig. 5-9. Transmitted IR power modulated by the GRIIRA effect in an APE MgO:LiNbO₃ waveguide. The IR and green light power density was 3.3MW/cm² and 0.31MW/cm² respectively.

Fig. 5-10 shows the measured absorption coefficient as a function of green light power density. The IR power density was fixed at 3.5MW/cm² in the tests. When the green light was changed from 0.12MW/cm² to 0.32MW/cm², the absorption coefficient of the none-APE MgO:LiNbO₃ waveguide increased from 2.2×10^{-2} /cm to 3.7×10^{-2} /cm, while the absorption coefficient of the APE MgO:LiNbO₃ waveguide increased from 2.7×10^{-2} /cm to 4.7×10^{-2} /cm. The APE MgO:LiNbO₃ demonstrated higher GRIIRA coefficients in the measured power range. The measured curve at over 0.2MW/cm² shows some degree of saturation. This trend is in agreement with absorption coefficient measurements on bulk MgO:LiNbO₃ crystals in literature, and is possibly due to depletion of excitable absorption centers. [5-19] [5-22]. The achievable green light power

density is limited by the output power of our green laser.

The absorption coefficient as a function of IR power was also measured. The results are shown in Fig. 5-11. Under 0.3MW/cm² green light power density, the absorption coefficient of both none-APE and APE MgO:LiNbO₃ waveguide gradually increased with increasing IR light power density. Such increase suggests that there were other mechanisms involved except the green light induced absorption.

The linear curve slope in Fig. 5-11 is approximately 6.7×10^{-3} /MW·cm for none-APE MgO:LiNbO₃, and 2.6×10^{-3} /MW·cm for APE MgO:LiNbO₃. In Fig. 5-10, before saturation at around 0.25MW/cm², the average linear curve slope is approximately 0.11/MW·cm for none-APE MgO:LiNbO₃, and 0.16/MW·cm for APE MgO:LiNbO₃. Therefore, the IR light self-induced absorption increase is only a few percent of the green light induced absorption, which is negligible in comparison. The possible origin of the IR self-induced absorption includes two photon absorption (TPA) [5-20], or excitation of shallow absorption centers through multiple absorption steps [5-23].

5-6. GRIIRA Induced Temperature and Refractive Index Modulations

Based on the measured GRIIRA coefficients, we are able to calculate the heat generation in the MgO:PPLN waveguide in the SHG process. As the absorption coefficient increases under higher green light power density, the generated heat also increases. Using simulation software, we are able to simulate the temperature distribution profile introduced by the GRIIRA effect.

Ph. D. Thesis – Jian Sun – Engineering Physics McMaster University



Fig. 5-10. GRIIRA absorption coefficient in MgO:LiNbO₃ sample (square) and the APE MgO:LiNbO₃ (circle) as a function of green light power density. The IR power density is fixed at 3.5MW/cm².



Fig. 5-11. GRIIRA absorption coefficient in MgO:LiNbO₃ sample (square) and the APE MgO:LiNbO₃ (circle) as a function of IR light power density. The green power density is fixed at 0.3MW/cm².

The simulation was carried out in COMSOL Multiphysics. First, a 3D heat transfer model was built. An MgO:LiNbO₃ ridge waveguide with 8 μ m width and 20 μ m height was set on a 500 μ m high substrate. The heat source was set at the light conducting area in the top part of the ridge. The environment was set at 20°C with natural convection. The bottom of the substrate was temperature-controlled at 20°C. Fig. 5-12 shows the model.

Using the data obtained in Fig. 5-10 and Fig. 5-11, we simulated the stationary temperature distributions in the waveguide. Under the light power density indicated in Fig. 5-10, the IR power in the waveguide was around 600mW, and the green light power was from about 20mW to 50mW. For Fig. 5-11, the green light power in the waveguide was approximately 50mW, and the IR power was from about 80mW to 600mW. A cross-sectional temperature profile of the waveguide was generated in each simulation, as shown in Fig. 5-13. The temperature increase in the waveguide's core was recorded, and the results are shown in Fig. 5-14.

From the simulation results in Fig. 5-14, we noticed that the waveguide core temperature increased 2.5°C under 600mW IR light and 50mW green light. Such temperature shift could significantly change the refractive index, and subsequently change the phase matching condition of the waveguide. Fig. 5-15 shows the refractive index change calculated from Fig. 5-14.

Since the absorption ratio is sensitive to the green light power, as the SHG process proceeds, the absorption ratio increases with the green light power. As a result, a temperature gradient is formed along the light propagation direction. Although the waveguide core temperature can be brought down by lowering the environment temperature, or changing the temperature controller, the temperature gradient cannot be eliminated with uniform temperature adjustments.



Fig. 5-12. (a) The 3D waveguide model used in the heat transfer simulation. (b) Cross section view of the model. The red area indicates the waveguide, which is the heat generating source.



Ph. D. Thesis – Jian Sun – Engineering Physics McMaster University

Fig. 5-13. Simulated stationary temperature distribution profile of MgO:LiNbO₃ waveguide. (a) and (b) are 3D and 2D cross-sectional view respectively. 78mW IR light power and 50mW green light power are used in the simulation.



Fig. 5-14. Waveguide core temperature increase caused by the GRIIRA effect. Simulation results in (a) and (b) are based on the results in Fig. 5-10 and Fig. 5-11 respectively.



Fig. 5-15. Waveguide refractive index change caused by the temperature modulation. The data is calculated using results in Fig. 5-13.

5-7. Influence of the GRIIRA Effect on SHG Performance

Using the obtained absorption data and the calculation method described in Chapter 2-2, we are able to build a simulation model for the MgO:PPLN waveguide to evaluate the potential impact of the GRIIRA effect on the SHG performance.

In the simulation, we assumed the waveguide is 8μ m wide, 20μ m high and 14mm long. The numbers were set to match the waveguide dimensions of simulations in Chapter 5-6 and experiments in Chapter 5-3. We used plane-wave approximation as described in Chapter 2-1. A mode overlap factor was added to adjust the conversion efficiency matching the experiment result shown in Fig. 5-4. The waveguide's conversion efficiency was normalized to 148%/W at 125mW IR power, which was shown in Fig. 5-4 at the lowest value. We consider at low IR input power, the measured conversion efficiency in was less affected by the GRIIRA effect. Using the finite difference simulation method described in Chapter. 2-1 and 2-2, we can build an SHG model without considering the SHG model, a small amount of phase mismatch was added to the Δk term in each step of iteration to simulate the temperature modulation introduced by GRIIRA effect. The amount of phase mismatch was proportional to the current generated green light power.

Fig. 5-16 shows the simulation result of the waveguide's power performance. We noticed that the waveguide's achievable output power was severely limited. The induced thermal effect limited the green light output under 150mW. The result suggests that under high power operation, improved thermal management method must be used on the waveguide to remove the heat produced by the GRIIRA effect. A single temperature controller on the back side of the crystal seems not sufficient, as the 500µm thick substrate produces a considerable amount of resistance.

The temperature-tuning curve of the waveguide was also simulated. Fig. 5-17 shows the curves under 100mW, 300mW, and 600mW IR light power. We noticed that the simulation results resemble the measurement results shown in Fig. 5-5. As the light power increases, the phase matching curve shifted to lower temperature, and became asymmetric. However, we were not able to reproduce the precise amount of shift as measured in Fig. 5-5. The temperature shift obtained through simulation was smaller than the measured values. Such result is expected, because the waveguide is also affected by other light-induced effect such as the photorefractive effect.

Although 5mol% MgO doped LiNbO₃ is much more resistant to photorefractive damage than none-MgO-doped LiNbO₃ [5-7]-[5-10]. It has been reported that the photorefractive effect is still present under kW/cm^2 IR light [5-7]. Under increased light power, the photorefractive effect increases the crystal's refractive index and dispersion, which in turn lowers the crystal's phase matching temperature. Although we are not able to quantitatively assess the impact of the photorefractive effect in the waveguide due to lack of data, based on empirical results, we believe that the refractive index modulation produced by the photorefractive effect is on a similar order of magnitude as produced by the GRIIRA effect. Because the SHG process in the waveguide is affected by multiple factors, and the amount of refractive index modulation depends on the generated green light itself, besides, the temperature distribution profile inside the waveguide is closely related to the waveguide's geometry, thermal management method and environmental conditions, it is very hard to precisely predict the performances of the waveguide under high input power. Although we are able to reproduce some of the tendencies observed in the SHG experiment, the simulation results in Fig. 5-16 and Fig. 5-17 are better to be treated as qualitative assessments as well.



Fig. 5-16. Simulated green light power generated in the MgO:PPLN waveguide with and without considering the GRIIRA effect.



Fig. 5-17. Simulated temperature tuning profiles of the waveguide under 100mW, 300mW, and 600mW input power. The GRIIRA effect is considered in the calculation.



Volume: Temperature (K)

Fig. 5-18. Simulated temperature distribution profile generated by the GRIIRA effect in MgO:LiNbO₃ waveguide. (a) The crystal is placed on a temperature controller with the waveguide facing upwards. (b) The waveguide is flip-mounted on a temperature controlled silicon substrate with adhesives.

There are several ways to reduce the impact of the GRIIRA effect, such as improving thermal management or changing the device design. Although it has been found that operating the device at an elevated temperature could help reduce the photorefractive effect [5-7], we did not find it helpful in reducing the GRIIRA effect in experiment. By increasing heat dissipation and reducing thermal resistance between the waveguide core and the temperature-controlled interface, we can reduce the temperature gradient and improve the performance of the waveguide. In Fig. 5-3, the waveguide is flip-mounted on a silicon substrate to improve thermal dissipation. We simulated the stationary temperature distribution of the structure using data in Fig. 5-10 and Fig. 5-11, and found that the temperature gradient dropped to about 20% of what shown in Fig. 5-14. Fig. 5-18 shows the temperature gradient generated with 50mW green light and 600mW IR light. Without the flip-mounting, the waveguide core temperature is 2.4°C higher than the environment temperature. In contrast, with the flip-mounting method, the waveguide core temperature is only 0.46 °C higher. The fact that we were able to generate around 450mW green light from the waveguide also approved that the thermal management method was effective.

5-8. Summary

In this chapter, we have generated 466 mW green light from an APE PPLN ridge waveguide with a conversion efficiency of around 70%. We have found significant refractive index change in the waveguide under increase light power, which is due to light induced effects such as photorefraction and GRIIRA. We have characterized the GRIIRA

coefficient in APE MgO:LiNbO₃ waveguides. To assess the impact of the GRIIRA effect on practical green lasers, simulations of stationary temperature distribution profiles have been performed. The results show considerable amount of temperature increase in the waveguide core under higher light power, which demands rigid thermal management. By applying the flip-mount design, the temperature gradient can reduce as much as 80%.

REFERENCES:

- [5-1]. Y. Enya, Y. Yoshizumi, T. Kyono, K. Akita, M. Ueno, M. Adachi, T. Sumitomo, S. Tokuyama, T. Ikegami, K. Katayama, and T. Nakamura, "531 nm green lasing of InGaN based laser diodes on semi-polar {2021} free-standing GaN substrates", Appl. Phys. Express 2, 082101 (2009).
- [5-2]. A. Tyagi, R. M. Farrell, K. M. Kelchner, C. Y. Huang, P. S. Hsu, D. A. Haeger, M. T. Hardy, C. Holder, K. Fujito, D. A. Cohen, H. Ohta, J. S. Speck, S. P. DenBaars, and S. Nakamura, "AlGaN-cladding freegreensemipolar GaN based laser diode with a lasing wavelength of 506.4 nm", Appl. Phys. Express 3, 011002 (2010).
- [5-3]. A. Avramescu, T. Lermer, J. Muller, C.Eichler, G. Bruederl, M. Sabathil, S. Lutgen, and U. Strauss, "True green laser diodes at 524 nm with 50 mW continuous wave output power on c-plane GaN", Appl. Phys. Express 3, 081003 (2010).
- [5-4]. K. Sakai, Y. Koyata, S. Itakura, and Y. Hirano, "High-Power, Highly Efficient Second-Harmonic Generation in a Periodically Poled MgO:LiNbO₃ Planar Waveguide", J. Lightwave Technol. 27, 590 (2009).
- [5-5]. K. Sakai, Y. Koyata, and Y. Hirano, "Planar-waveguide quasi-phase-matched

second-harmonic-generation device in Y-cut MgO-doped LiNbO₃", Opt. Lett. 31, 3134 (2006).

- [5-6]. H. K. Nguyen, M. H. Hu, Y. Li, K. Song, N. J. Visovsky, S. Coleman, and C. E. Zah, Optical Components and Materials V, Proc. of SPIE 6890, 689001 (2008).
- [5-7]. M. Asobe, O. Tadanaga, T. Yanagawa, H. Itoh, and H. Suzuki, "Reducing photorefractive effect in periodically poled ZnO- and MgO- doped LiNbO₃ wavelength converters", Appl. Phys. Lett. 78, 3163 (2001).
- [5-8]. Y. Furukawa, K. Kitamura, A. Alexandrovski, R. K. Route, M. M. Fejer, and G. Foulon, "Green-induced infrared absorption in MgO doped LiNbO₃", Appl. Phys. Lett. 78, 1970 (2001).
- [5-9]. Y. Furukawa, K. Kitamura, S. Takekawa, A. Miyamoto, M. Terao, and N. Suda, "Photorefraction in LiNbO₃ as a function of [Li]/[Nb] and MgO concentrations", Appl. Phys. Lett. 77, 2494 (2000).
- [5-10]. S. Kan, M. Sakamoto, Y. Okano, D. H. Yoon, T. Fukuda, O. Oguri, and T. Sasaki, "Photorefraction Dependent on Composition in LiNbO₃ Crystals with and without MgO Doping", Cryst. Res. Technol. 31, 353 (2006).
- [5-11]. M. Simon, F. Jermann, and E. Kratzig, "Light-induced absorption changes in iron-doped LiNbO₃", Opt. Mater. 3, 243 (1994).
- [5-12]. F. Jermann, M. Simon, and E. Kratzig, "Photorefractive properties of congruent and stoichiometric lithium niobate at high light intensities", J. Opt. Soc. Am. B 12, 2066 (1995).
- [5-13]. K. Buse, F. Jermann, and E. Kratzig, "Infrared holographic recording in

LiNbO₃:Fe and LiNbO₃:Cu", Opt. Mater. 4, 237 (1995).

- [5-14]. J. Hirohashi, V. Pasiskevicius, S. Wang, and F. Laurell, "Picosecond blue-light-induced infrared absorption in single-domain and periodically poled ferroelectrics", J. Appl. Phys. 101, 033105 (2007).
- [5-15]. P. Herth, T. Granzow, D. Schaniel, Th. Woike, M. Imlau, and E. Kratzig, "Evidence for light-induced hole polarons in LiNbO₃", Phys. Rev. Lett. 95, 067404 (2005).
- [5-16]. F. Luedtke, K. Buse, and B. Sturman, "Hidden reservoir of photoactive electrons in LiNbO₃ crystals", Phys. Rev. Lett. 109, 026603 (2012).
- [5-17]. N. Waasem, A. Markosyan, M. M. Fejer, and K. Buse, "Green-induced blue absorption in MgO-doped lithium niobate crystals", Opt. Lett. 38, 2953 (2013).
- [5-18]. M. Iwai, T. Yoshino, S. Yamaguchi, M. Imaeda, N. Pavel, I. Shoji, and T. Taira, "High-power blue generation from a periodically poled MgO:LiNbO₃ ridge-type waveguide by frequency doubling of a diode end-pumped Nd:Y₃Al₅O₁₂ laser", Appl. Phys. Lett. 83, 3659 (2003).
- [5-19]. J. Hirohashi, T. Tago, O. Nakamura, A. Miyamoto, and Y. Furukawa, "Characterization of GRIIRA properties in LiNbO₃ and LiTaO₃ with different compositions and doping", Proc. of SPIE 6875, 687516-1 (2008).
- [5-20]. A. Carson and M. E. Anderson, "Two-photon absorption and blue-light-induced red absorption in LiTaO₃ waveguides", JOSA B 23, 1129 (2006).
- [5-21]. D. Berben, K. Buse, S. Wevering, P. Herth, M. Imlau, and Th. Woike, "Lifetime of small polarons in iron-doped lithium–niobate crystals", J. Appl. Phys, 87, 1034

(2000).

- [5-22]. J. Hirohashi, V. Pasiskevicius, S. Wang, and F. Laurell, "Picosecond blue-light-induced infrared absorption in single-domain and periodically poled ferroelectrics", J. Appl. Phys. 101, 033105 (2007).
- [5-23]. N. Waasem, A. Markosyan, M. M. Fejer, and K. Buse, "Green-induced blue absorption in MgO-doped lithium niobate crystals", Opt. Lett. 38, 2953 (2013).

Chapter 6. 1.5-µm Communication Band Second Harmonic Generation

6-1.Application Background

PPLN waveguides have been widely adopted in 1.5µm-band optical communication devices for high efficiency wavelength conversion. SHG, DFG, or cascade second-order nonlinear optical conversion has been demonstrated in various applications such as parametric amplifiers and frequency mixers [6-1]-[6-6]. Today, 5mol% MgO doped LiNbO₃ are commonly used instead of the none-doped crystal, because of its higher resistance to optical damages at high power levels [6-7]-[6-9].

Recently, PPLN ridge waveguides have been reported with higher conversion efficiency [6-5] [6-10] than traditional buried waveguides, because of the improved light confinement factor resulting from higher refractive index contrast. PPLN waveguides have also been adopted in generating entangled photon pairs used in communication band optical quantum teleportation [6-11]-[6-15]. The low propagation loss and high nonlinear coefficient ensures efficient amplification from a single photon in the phase-matched bandwidth.

In this chapter, we demonstrate efficient 780nm-band SHG from 1560nm-band using an APE MgO:PPLN waveguide. Using a tunable laser and a temperature controlling unit, wavelength tuning can be easily achieved. The generated tunable 780nm-band light can be used as a flexible pump source for second stage nonlinear conversions or photon pair generations.

6-2. Device Design and Fabrication

The waveguide was designed to be integrated with 1.5µm-band polarization maintaining (PM) optical fiber, which had a mode field diameter of 9.9µm. To provide good coupling efficiencies with fibers, the APE MgO:PPLN ridge waveguide was designed to be 10µm wide. The proton-exchange condition was set to 2.5 hours at 200°C and the annealing condition is 6 hours at 350°C. Fig. 6-1 shows the calculated refractive index profile generated by proton-exchange.

The 1.5µm-band tunable laser used in this experiment was composed of a seeding laser and an amplifier. The output range was from 1520nm to 1560nm. To avoid back reflections from the waveguide facets and transmission fluctuations caused by the Fabry-Perot effect, the waveguide facets were cut to a 9° angle. Ridge waveguides with 10µm and 12µm were fabricated. The length of the waveguides was 19mm. Fig. 6-2 shows photos of the waveguide.



Fig. 6-1. Calculated refractive index profile for APE MgO:LiNbO₃ waveguide at (a) 1560nm, and (b). 780nm. The waveguide is processed with 2.5 hours of PE and 6 hours of annealing time.

Ph. D. Thesis – Jian Sun – Engineering Physics McMaster University



Fig. 6-2. Microscope photos of the fabricated ridge waveguides. The waveguide facets are cut to 9° angle to prevent back-reflection. The chip contains a 10µm wide and a 12µm ridge waveguide.
6-3. Experiment Result

After fabrication, the waveguide was first tested using a 1.5µm-band broadband source. Light was coupled into and out of the ridge waveguides using single mode optical fibers. The SHG signals from the waveguide was collected and measured in a spectrum analyzer. Fig. 6-3 shows the measured spectrum of a 12µm wide and 10µm wide waveguide. At room temperature, the phase-matching wavelength for the 10µm and 12µm wide waveguide was 780.0nm 785.6nm respectively. The full width half maximum (FWHM) phase matching bandwidth for the waveguide was 0.34nm and 0.45nm respectively. The asymmetric side lopes in the figure may come from imperfect poling or small fluctuations of waveguide width.

The fundamental wavelength for the 785.6nm SHG signal is 1571.2nm, which was beyond of the range of our tunable laser. The phase matching wavelength of the 10µm wide waveguide can be tuned to a shorter wavelength by lowering the crystal's temperature. However, we found it would require turning the temperature controller to below zero degree, which cannot be done at room condition due to frosting issues. The phase matching wavelength can also be tuned by changing the poling period of the PPLN crystal. However, due to availability of our PPLN crystals, the 12µm wide waveguide was used in the following SHG experiment.

In the SHG experiment, the waveguide was placed on a temperature controlled optical stage. The 1.5µm-band tunable laser was coupled into the waveguide using a PM fiber. Since the APE waveguide only supports TM polarization, the PM fiber was physically rotated to TM position to obtain maximum coupling efficiency. The output light was collected by an objective lens, filtered, and measured by an optical power meter.

Ph. D. Thesis – Jian Sun – Engineering Physics McMaster University



Fig. 6-3. Normalized SHG spectrum of (a) 12μm wide and (b) 10μm wide ridge waveguide. The spectrum is measured using a 1.5μm-band broadband light source.

The measured insertion loss of the waveguide was around 4 dB, which was contributed by the waveguide's coupling loss, Fresnel reflection loss, and propagation loss. A maximum output of 4.3mW 780nm light was received at the power meter. By considering the Fresnel loss at the output facet, and transmission rate of the filter, we calculated that 4.9mW of 780nm light was generated in the waveguide under 110mW of 1560nm light. Fig.6-4 shows the generated SHG light power at different fundamental light power. We calculate the conversion efficiency of the waveguide was 4.45% or normalized efficiency $11.2\%/(W \cdot cm^2)$.



Fig. 6-4. Power performance of the 12μm wide 19mm long waveguide. A maximum power of 4.3mW 780nm light is generated with 110mW 1560nm light.

The efficiency of the waveguide is limited by two main factors, the waveguide length and waveguide width. Our current 19mm waveguide length is limited by the available length of poling sections of the PPLN crystal. The 12µm waveguide width is limited by the available poling period. By using a longer crystal with slightly shorter poling period, the waveguide's performance can be largely enhanced. Based on the data obtained with our current 12µm wide 19mm long waveguide, we calculate that for a 45mm long 10µm wide waveguide, the conversion efficiency under 110mW of 1560nm light would be approximately $11.2\%/(W \cdot cm^2)*(4.5cm)^{2*}(12µm/10µm)*0.11W=30\%$. By further increasing the 1560nm light power, even higher conversion efficiency is achievable.

6-4. Summary

In this chapter, we have demonstrated efficient 780nm light generation using APE MgO:LiNbO₃ waveguide. The normalized conversion efficiency is $11.2\%/(W \cdot cm^2)$. The conversion efficiency can be enhanced by increasing the waveguide length and reducing the ridge width. We estimate that 30% conversion efficiency can be achieved using a 45mm long, 10µm wide waveguide with 110mW pump light.

REFERENCES:

- [6-1]. K. R. Parameswaran, R. K. Route, J. R. Kurz, R. V. Roussev, M. M. Fejer, and M. Fujimura, "Highly efficient second-harmonic generation in buried waveguides formed by annealed and reverse proton exchange in periodically poled lithium niobate", Opt. Lett. 27, 179 (2002).
- [6-2]. K. R. Parameswaran, J. R. Kurz, R. V. Roussev, and M. M. Fejer, "Observation of 99% pump depletion in single-pass second-harmonic generation in a periodically

poled lithium niobate waveguide", Opt. Lett. 27, 43 (2002).

- [6-3]. M. H. Chou, J. Hauden, M. A. Arbore, and M. M. Fejer, "1.5-mm-band wavelength conversion based on difference-frequency generation in LiNbO₃ waveguides with integrated coupling structures", Opt. Lett. 23, 1004 (1998).
- [6-4]. I. Brener, M. H. Chou, E. Chaban, K. R. Parameswaran, M. M. Fejer, S. Kosinski, and D. L. Pruitt, "Polarisation-insensitive wavelength converter based on cascaded nonlinearities in LiNbO₃ waveguides", Electron. Lett. 36, 66 (2000).
- [6-5]. Y. Nishida, H. Miyazawa, M. Asobe, O. Tadanaga, and H. Suzuki, "0-dB wavelength conversion using direct-bonded QPM-Zn:LiNbO₃ ridge waveguide", Photon. Technol. Lett. 17, 1049 (2005).
- [6-6]. A. Galvanauskas, K. K.Wong, K. El Hadi, M. Hofer, M. E. Fermann, D. Harter, M. H. Chou, and M. M. Fejer, "Amplification in 1.2-1.7 μm communication window using OPA in PPLN waveguides", Electron. Lett. 35, 731 (1999).
- [6-7]. Y. Furukawa, K. Kitamura, S. Takekawa, A. Miyamoto, M. Terao, and N. Suda, "Photorefraction in LiNbO₃ as a function of [Li]/[Nb] and MgO concentrations", Appl. Phys. Lett. 77, 2494 (2000).
- [6-8]. Y. Furukawa, K. Kitamura, A. Alexandrovski, R. K. Route, M. M. Fejer, and G. Foulon, "Green-induced infrared absorption in MgO doped LiNbO₃", Appl. Phys. Lett. 78, 1970 (2001).
- [6-9]. M. Asobe, O. Tadanaga, T. Yanagawa, H. Itoh, and H. Suzuki, "Reducing photorefractive effect in periodically poled ZnO- and MgO-doped LiNbO₃ wavelength converters", Appl. Phys. Lett. 78, 3163 (2001).

- [6-10]. T. Kishimoto and K. Nakamura, "Periodically poled MgO-doped stoichiometric LiNbO₃ wavelength converter with ridge-type annealed proton-exchanged waveguide", Photon. Technol. Lett. 23, 161 (2011).
- [6-11]. S. Arahira, N. Namekata, T. Kishimoto, H. Yaegashi, and S. Inoue, "Generation of polarization entangled photon pairs at telecommunication wavelength using cascaded $\chi^{(2)}$ processes in a periodically poled LiNbO₃ ridge waveguide," Opt. Express 19, 16032 (2011).
- [6-12]. H. C. Lim, A. Yoshizawa, H. Tsuchida, and K. Kikuchi, "Stable source of high quality telecom-band polarization-entangled photon-pairs based on a single, pulse-pumped, short PPLN waveguide," Opt. Express 16, 12460 (2008).
- [6-13]. H. C. Lim, A. Yoshizawa, H. Tsuchida, and K. Kikuchi, "Stable source of high quality telecom-band polarization-entangled photon-pairs based on a single, pulse-pumped, short PPLN waveguide," Opt. Express 16, 12460 (2008).
- [6-14]. S. Arahira and H. Murai, "Nearly degenerate wavelength-multiplexed polarization entanglement by cascaded optical nonlinearities in a PPLN ridge waveguide device," Opt. Express 21, 7841 (2013).
- [6-15]. S. Arahira, N. Namekata, T. Kishimoto, and S. Inoue, "Experimental studies in generation of high-purity photon-pairs using cascaded $\chi^{(2)}$ processes in a periodically poled LiNbO₃ ridge-waveguide device," J. Opt. Soc. Am. B 29, 434 (2012).

Chapter 7. 3.3 µm-Band Mid-Infrared Difference Frequency Generation

7-1. Application Background

Compact low-cost mid-infrared (mid-IR) lasers are highly demanded in various kinds of applications such as high-resolution spectroscopy, trace gas detection and remote sensing. In spectroscopy applications, mid-IR laser with milliwatt level output is desired for obtaining satisfactory performances. Among the reported mid-IR laser technologies, difference frequency generation (DFG) via second-order nonlinear wavelength conversion is a popular method due to its simplicity, wide tunability and room temperature operation. LiNbO₃ crystals have been widely accepted for 2~5µm mid-IR generation due to its good transparency and large second-order nonlinear coefficient.

DFG mid-IR laser using a PPLN waveguide is very promising because of its high conversion efficiency and the ability of integration with semiconductor laser and fiber systems. In recent years, efficient mid-IR light generation using wafer bonded PPLN waveguide has been reported. O. Tadanaga *et al.* [7-1] demonstrated DFG at 3µm wavelength range using 1µm and 1.5µm band lasers in a 50mm long LiNbO₃ ridge waveguide. 0.26mW mid-IR output was obtained. M. Asobe *et al.* [7-2] reported 3.4µm mid-IR DFG using a 1064nm pump and 1550nm signal laser. The output reached 65mW under a signal power of 558mW and a pump power of 444mW. T. Yanagawa *et al.* [7-3] showed measurement of methane absorption spectrum using mid-IR light generated using tunable laser diodes and a PPLN ridge waveguide.

In this chapter, we demonstrate milliwatt-level mid-IR generation using our APE MgO:PPLN ridge waveguide. Using a dual-wavelength high power fiber laser emitting at

1057.7nm and 1554.3nm, 3.3μ m band mid-IR light is generated through quasi phase matched DFG in the waveguide. A maximum output of 2mW is obtained. Using a 1064nm laser and a 1.5 μ m band tunable laser, tunable mid-IR light ranging from 3.3μ m to 3.5μ m can be generated. Mid-IR light at this wavelength range can be used in combustible gas detection such as methane.

7-2. Waveguide design and fabrication

We use a dual-wavelength, linearly polarized fiber laser as our pump laser. The laser was kindly provided by Dr. Xijia Gu at Ryerson University. The fiber laser emits 1060nm-band and 1550nm-band light simultaneously, which can be coupled into the ridge waveguide through a PM fiber. We measured the spectrum of the laser, and the results are shown in Fig. 7-1. We found that the 1060nm-band output was centered at 1057.2nm, and the 1550nm-band output was centered at 1554.0nm. The full width half maximum (FWHM) line width was 63pm and 48pm respectively. The actual 1554nm line might be narrower because of the spectrum analyzer's 50pm resolution.

The mode field diameter of the PM fiber used in the dual-wavelength fiber laser is $9.9\mu m$ at 1550nm-band. To ensure good coupling from the fiber to the waveguide, the ridge width should be larger than 10 μm . The generated mid-IR light is at 3310nm. To calculate the refractive index profile of the waveguide, we need to know the refractive index increment at this wavelength after proton-exchange. However, we are unable to find any literature on this point. To obtain a satisfactory result, we borrowed the empirical results from several reported mid-IR DFG using APE channel waveguides [7-4] [7-5], and tried to find out the optimized fabrication conditions experimentally.



Fig. 7-1. Spectrum of the dual wavelength fiber laser. The output wavelengths are centered at 1057.2nm and 1554.0nm, with FWHM bandwidth of 63pm and 48pm respectively.

We fabricated APE MgO:PPLN ridge waveguide with 8~16 hours of PE time and various annealing time. The ridge widths were 10, 12, and 16µm, and the waveguide length was 38mm. After each fabrication, the waveguide was tested using methods described in the following chapter 7-3 to find out the phase matching conditions. From chapter 3, Fig. 3-13, we have learned that the a certain amount of annealing time is needed to convert phase *b* MgO:H_xLi_{1-x}NbO₃ crystals to phase *a* and phase *c*. However, we were unable to find clear phase matched signal from the well-annealed samples containing only phase *a* and phase *c* crystals. One possible reason is due to reduced refractive index contrast at longer annealing time, which results in mode cut-off at the mid-IR wavelength. To solve this issue, we made a trade-off between refractive index contrast and crystal recovery by using shorter annealing time. However, the drawback of the trade-off is increased absorption loss introduced by the residue phase *b* crystals, which possess a broadband absorption tail extending beyond 3450cm⁻¹ (>2.9µm).

7-3. Characterization of Phase Matching Conditions by SPDC Spectroscopy

After fabrication, the ridge waveguides were tested using the spontaneous parametric down conversion (SPDC) spectroscopy method. We used a high power 1064nm laser as the pumping source. Through the SPDC process, a 1064nm photon was split into a 1550nm band and a 3310nm band photon, which was determined by the phase matching condition of the PPLN crystal. With Watt-level pumping power, the SPDC signal was amplified strong enough to be detected using spectrum analyzer. Fig. 7-2 shows the experiment setups for the tests. Around 1W of 1064nm pump light was coupled into the

waveguide using PM fiber. The output light from the waveguide was collected by an objective lens. The light was filtered by a 1200nm long-pass filter to block the pump light before entering the spectrum analyzer. The 1550nm band signal was analyzed and recorded.



Fig. 7-2. Experiment setup of the SPDC spectrum test. A high power 1064nm fiber laser is used to couple over 1W of light into the waveguide. The 1550nm-band SPDC signal is collected, filtered, and measured in the spectrum analyzer.

Fig. 7-3 shows the obtained spectrum from a 12µm wide and 16µm wide waveguides with a poling period of 29.5µm. The 12µm wide waveguide shows a phase matching signal at 1535nm, while the 16µm wide waveguide is phase matched at 1501nm. The results were obtained at room temperature of 24°C. The APE condition was 12 hours of PE at 200°C and 12 hours of annealing at 350°C. We were unable to obtain any signal from the 10µm wide waveguide. We consider the mid-IR light reached cut-off in narrower waveguides.

In order to move the phase matching wavelength to around 1560nm, we need to increase the crystal's temperature or increase the crystal's poling period. We fabricated

12μm wide waveguide on another PPLN crystal with 30μm period. The measured spectrum is shown in Fig. 7-4. However, we found that the phase matching wavelength was 1573.2nm, which was beyond 1560nm. As a result, we decided to continue to use the 12μm wide waveguide with 29.5μm poling period. We measured the temperature tuning characteristics of the waveguide. In Fig. 7-5, we found that the phase matching wavelength could be changed to 1541.6nm at 45 °C, and to 1550.6nm at 65 °C. The temperature tunability of the waveguide was around 0.4nm/ °C at 1550nm-band, which correspond to 1.9 nm/ °C at 3310nm-band. We estimate the phase matching wavelength can reach 1560nm-band at around 85 °C.



Fig.7-3 SPDC spectrum of a 12µm wide waveguide (right) and a 16µm wide waveguide (left).



Fig. 7-4. SPDC spectrum of a12µm wide waveguide with 29.5µm (left) and 30µm (right) poling period.



Fig. 7-5. SPDC spectrum of a 12μm wide waveguide measured at 25°C (1535nm), 45 °C (1541.6nm), and 65 °C (1550.6nm).

7-4. Milliwatt-Level Mid-IR Generation

To test the power performance, the waveguide was tested using setups shown in Fig. 7-6. The waveguide was placed on a temperature controlled stage with 0.1 °C accuracy. Light from the dual wavelength fiber laser was coupled into the waveguide by PM fiber. The PM fiber was rotated to align with the waveguide at TM polarization. We did not observe any power fluctuation caused by the F-P effect due to the broad line width of the laser.

The output light from the waveguide was collected using a CaF_2 lens. The light was filtered by a 3mm thick Ge filter, which cut off any visible or near-IR light shorter than 1800nm wavelength. The mid-IR light was measured by a stabilized thermal detecting optical power meter with a sensitivity of 1µW.

We also measured the 1057nm and 1554nm output power from the waveguide individually using corresponding near infrared filters. Considering the transmission rates of the lens, filter, and the Fresnel reflection loss of the waveguide's output facet, we were able to calculate the power of mid-IR light generated by the waveguide. The values were recorded as a function of the dual-wavelength fiber laser's pump current, which were shown in Fig. 7-7 and Fig. 7-8.

A maximum of 226mW 1057.2nm light and 283mW of 1554.0nm light was coupled into the waveguide, while the dual wavelength fiber laser was working under 8A of pump current. A maximum of 1.6mW 3307nm light was obtained. The phase matching temperature of the waveguide was 80°C. In Fig. 7-8, the mid-IR output power shows a potential for further increasing. However, due to power limitations of our dual

wavelength fiber laser, we were unable to obtain higher mid-IR power. By replacing the dual wavelength fiber laser with a 1064nm laser and a 1.5μ m-band tunable laser, a tunable mid-IR laser in 3.3μ m to 3.5μ m-band can be easily built.



Fig.7-6. Experiment setup for milliwatt-level mid-IR light generation.



Fig. 7-7. 1057nm and 1554nm light power coupled into the waveguide. The horizontal axis is the pump current of the dual wavelength fiber laser.



Fig. 7-8. Mid-IR light power generated by the waveguide. The horizontal axis is the pump current of the dual wavelength fiber laser.

We calculated the conversion efficiency of the APE MgO:PPLN waveguide, and found that the value of 2.5%/W is much lower than that reported in wafer-bonded waveguide [7-1]. We consider the reason is two-fold.

The first reason is due to weak confinement of the mid-IR light. From our experiment, we observed that well-annealed APE waveguides satisfying the relationship found in Fig. 3-13 were not able to produce clear SPDC signals. We also observed that in 10µm wide waveguide, the mid-IR light possibly reached cut-off. Because of weak light confinement, mode overlap among 1057nm, 1554nm and 3307nm light was poor, which in turn reduced the conversion efficiency.

The second reason is due to absorption loss introduced by the remaining phase *b* crystals. We have to make a trade-off between light confinement and crystal recovery. According to the spectrum study in chapter 3, the remaining phase *b* crystals possess an absorption tail expanding to over 3μ m. We have re-examined the FTIR transmission spectrum of APE MgO:LiNbO₃ with 12 hours of PE time and 12 hours of annealing time, and compared it with none-processed MgO:LiNbO₃. The result is shown in Fig. 7-9. We found there was a slight increase of 0.3% absorption at 3.3µm band. The FTIR spectrum was measured with beam direction perpendicular to the crystal wafer, and according to our calculation, the APE layer that the light traveled through was only around 20µm thick. According to these numbers, we estimate that the propagation loss for 3.3µm light in the waveguide is about 0.7dB/mm, which could lead to severe performance degradation in a 38mm long waveguide.

However, despite the lower than expected conversion efficiency, we are still able to obtain mid-IR light with milliwatt-level output power, which is sufficient for many gas detection and spectroscopy applications.



Fig. 7-9. FTIR spectrum of bulk MgO:LiNbO₃ crystal and APE MgO:LiNbO₃ crystal with 12 hours of PE and 12 hours of annealing. The beam direction is perpendicular to the crystal surface with APE. The transmission difference at 3.3µm wavelength is around 0.3%.

7-5. Summary

In this chapter, we have demonstrated $3.3\mu m$ light generation via DFG of 1057nm and 1554nm light in an APE MgO:LiNbO₃ ridge waveguide. Although the waveguide's performance is limited by additional absorptions introduced by the OH⁻ groups, we are able to obtain 1.6mW output power. The tunability of the waveguide is approximately 1.9 nm/°C at the $3.3\mu m$ -band.

REFERENCES:

- [7-1]. O. Tadanaga, T. Yanagawa, Y. Nishida, H. Miyazawa, K. Magari, M. Asobe, and H. Suzuki, "Efficient 3-μm difference frequency generation using direct-bonded quasi-phase-matched LiNbO₃ ridge waveguides", Appl. Phys. Lett. 88, 061101 (2006).
- [7-2]. M. Asobe, O. Tadanaga, T. Yanagawa, T. Umeki, Y. Nishida and H. Suzuki, "High-power mid-infrared wavelength generation using difference frequency generation in damage-resistant Zn:LiNbO₃ waveguide", Electron. Lett. 44, No.4 (2008).
- [7-3]. T. Yanagawa, O. Tadanaga, K. Magari, Y. Nishida, H. Miyazawa, M. Asobe, and H. Suzuki, "CH₄ monitoring in ambient air by communication band laser diode based difference frequency generation in a quasi-phase-matched LiNbO₃ waveguide", Appl. Phys. Lett. 89, 221115 (2006).
- [7-4]. K.P. Petrov, A.P. Roth, T.L. Patterson, T.P.S. Thoms, L. Huang, A.T. Ryan, and D.J. Bamford, "Efficient difference-frequency mixing of diode lasers in lithium niobate channel waveguides", Appl. Phys. B. 70, 777 (2000).
- [7-5]. R. V. Roussev, S. Sinha, R. L. Byer, and M. M. Fejer, "Efficient Mid-IR Generation in Reverse-Proton Exchanged Lithium Niobate Waveguides", Frontiers in Optics, OSA Technical Digest, paper JWD46 (2006).

Chapter 8. Conclusions

Annealed proton-exchanged MgO:PPLN ridge waveguides have been successfully developed. Using the well-established APE techniques and the state-of-the-art precise machining techniques, ridge waveguides with average surface roughness less than 4nm, and good uniformity over 5cm length have been successfully achieved. The waveguide propagation loss was measured around 0.2dB/cm at near-IR wavelengths. The process has shown good repeatability and flexibility during research. Comparing to other types of LiNbO₃ waveguides, the APE ridge waveguide combines flexibility, repeatability, high conversion efficiency, and low fabrication cost.

A simulation model for the APE MgO:PPLN ridge waveguide has been built. Using data from literatures and experiments, the refractive index profile of the waveguide can be simulated. The simulated mode profile matched well with experiment measurement.

Using FTIR spectroscopy, phase transitions in APE MgO:LiNbO₃ have been studied and compared to none-doped APE LiNbO₃. The proton diffusion constant of APE MgO:LiNbO₃ during proton-exchange was calculated. The FTIR spectrum of APE MgO:LiNbO₃ revealed three independent absorption peaks located in the OH stretching vibration band, which suggested that the proton ions in the crystal were in at least three different phases. Phase *a* 3479cm⁻¹, phase *b* 3501cm⁻¹, phase *c* 3533cm⁻¹. The evolution of FTIR spectrum during annealing showed that the proton concentration relationship was phase *b* > phase *a* > phase *c*. Measurement of waveguide propagation loss and phase matching wavelengths showed that the second-order nonlinear coefficient d_{33} is near zero in phase *b*, while non-zero in phase *a* and phase *c*. The waveguide reached optimized performance when phase *b* was depleted and transformed into phase *a* and phase *c* after

annealing. The obtained result can be used in optimizing the proton-exchange and annealing times during fabrication.

466mW of 532nm green light has been generated using a single APE MgO:PPLN ridge waveguide. The ridge waveguide was 8µm wide and 14mm long. Through second harmonic generation (SHG) from a 1064nm fiber laser, we obtained an optical to optical conversion efficiency of 69.7%. Heat generation in the waveguide has been observed under high power operation. The green induced infrared absorption (GRIIRA) effect has been studied. Absorption coefficient has been measured under various conditions. Using the measured data, temperature distribution profiles in the waveguide were simulated. The temperature gradient introduced refractive index modulation in the waveguide, which changed the phase matching conditions of the waveguide. Through simulations, it was found that the GRIIRA effect can significantly limit the performance of MgO:PPLN waveguides. Using proper thermal management method, the temperature gradient brought by the GRIIRA effect can be reduced. Simulation results showed that by flip-mount the ridge waveguide on a silicon substrate, the temperature gradient between the waveguide core and the environment can be reduced by over 80%.

780nm SHG from 1560nm communication band laser has been demonstrated using the APE MgO:PPLN ridge waveguide. A tunable 1.5μm-band laser was used in the experiment. 4.9mW of 780nm light was generated under 110mW of 1560nm light in a 12μm wide 19mm long waveguide. The waveguide has great potentials for further optimization. Using PPLN crystals with shorter poling period, calculations have shown that a 10μm wide 40mm long waveguide can easily deliver over 100mW of 780nm light. The generated tunable780nm-band light can be used as a pump source for optical parametric generations or photon pair generations.

A 3.3µm-band mid-IR laser has been built with the APE MgO:PPLN ridge waveguide. Through difference frequency generation (DFG) in a 12µm wide 36mm long waveguide, 1.6mW of 3307nm mid-IR light was generated using a dual wavelength fiber laser operating at 1057.2nm and 1554.0nm. The phase matching conditions and temperature tuning characteristics of the waveguide have been studied using spontaneous parametric down conversion (SPDC) spectroscopy. By combining the waveguide with a 1064nm laser and a 1.5µm-band tunable laser, a 3.3µm-3.5µm band tunable mid-IR laser can be built. Although several factors have been found limiting the waveguide's performance, the generated milliwatt-level mid-IR light is sufficient for gas detection and spectroscopy applications.

In conclusion, the APE MgO:PPLN ridge waveguide has been successfully developed, and its optical properties have been researched. Light generation experiments have been successfully conducted using the waveguide. The APE MgO:PPLN ridge waveguide has been proved suitable for high efficiency nonlinear optical conversions across its transparency spectrum from visible to mid-IR light. With distinctive features including flexibility, high efficiency and low cost, this new type of waveguide can find potential applications in various fields for wavelength conversion purposes. As the frontier of optics expands, it could also open up more possibilities in future researches.

Appendix

FTIR spectrum of APE MgO:LiNbO3 crystals with various PE and annealing time









List of Publications

J. Sun and C. Q. Xu, "Correlation of Infrared Spectra and Phase Transitions in Annealed Proton-Exchanged MgO doped LiNbO₃", submitted to J. of Appl. Phys., 2014.

J. Sun and C. Q. Xu, "Green Induced Infrared Absorption in Annealed Proton-Exchanged MgO:LiNbO₃ Waveguides", submitted to JOSA B, 2014.

A. R. EL-Damak, J. H. Chang, <u>J. Sun</u>, C. Q. Xu, and X. Gu, "Dual-Wavelength, Linearly Polarized All-Fiber Laser With High Extinction Ratio", IEEE Photonics Journal 5, 1501406 (2013).

J. Sun and C.Q. Xu, "466 mW Green Light Generation using Annealed Proton Exchanged Periodically Poled MgO:LiNbO₃ Ridge Waveguides", Opt. Lett. 37, 2028 (2012).

Y. Gan, Q. Y. Xu, Y. Lu, <u>J. Sun</u>, and C. Q. Xu, "Watt-level compact green laser for projection display", Journal of the Society for Information Display 19, 833 (2012).

C. Q. Xu, Y. Gan, and <u>J. Sun</u>, "MgO:PPLN frequency doubling optical chips for green light generation: from lab research to mass production", Proc. SPIE 8280, (2012).

J. Sun and C. Q. Xu, "Efficient green-light generation by proton-exchanged periodically poled MgO:LiNbO₃ ridge waveguide", Opt. Lett. 36, 549 (2011).

C. Q. Xu, <u>J. Sun</u>, and Y. Gan, "Green light generation based on periodically poled LiNbO₃ waveguides", Proc. SPIE 8007, (2011).