ANNEALED PROTON-EXCHANGED LiNbO₃ RIDGE WAVEGUIDES -- SIMULATION, FABRICATION AND CHARACTERIZATION

BY

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B.Sc
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TITLE: Annealed Proton-Exchanged LiNbO3 Ridge Waveguides -- Simulation, Fabrication and Characterization

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Abstract

Although semiconductor optical waveguides are quite mature in industry productions, fabrication of high quality lithium niobate (LiNbO$_3$) waveguides has always been a challenging topic over the past decades. Intense studies have been done, and various kinds of fabrication methods have been invented, however, existing technologies still cannot provide a satisfying solution with both excellent performances and easy processing. Traditional ion doped optical waveguides have poor light confinement, while the newly emerged etching and precise machining techniques are still under developing. Searching for new fabrication methods for LiNbO$_3$ waveguide devices is a meaningful pursuit of both academic and practical value.

In this study, a novel fabrication technique for LiNbO$_3$ waveguides has been proposed and realized. Through the combination of proton-exchange and precise diamond blade dicing, the annealed proton-exchanged (APE) LiNbO$_3$ ridge waveguides with low propagation loss and single transmission mode have been fabricated for both 1550 nm and 1064 nm wavelengths. The results have shown good agreements with theoretical simulation. Based on the obtained data, a nonlinear second harmonic generation (SHG) wavelength converter has been fabricated using the APE ridge waveguide built on periodically polled MgO:LiNbO$_3$ (PPMgLN) substrate. Test results have shown remarkable improvements comparing with traditional waveguide structures.

Although there are remaining problems need to be solved, the fabricated waveguides have demonstrated great potentials for various optical applications. Based on this platform, mid-infrared generation by difference frequency generation (DFG), compact blue laser via SHG, high-speed optical modulators and many other useful devices can be built. Not all of them have been fully exploited yet.
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Chapter 1. Introduction

1-1. General Property of LiNbO$_3$ Crystal

LiNbO$_3$ (Lithium Niobate) crystal is one of the extensively used material in scientific researches and applications. It possesses many extinguishing characteristics such as ferroelectricity, piezoelectricity, large electro-optic and acousto-optic coefficient, very broad transmission band, and high nonlinear optical coefficient at the same time. By tailoring its properties, various useful devices can be made from LiNbO$_3$ crystal.

LiNbO$_3$ does not exist in nature, in other words, it’s an artificial crystal, which is a typical successful case of modern crystal synthesis technology. Industrial production method for LiNbO$_3$ crystal is based on the well-know Czochralski process. However, the crystal grown under congruent condition presents large concentration of intrinsic defects, and these defect sites can be altered by doping different impurities into the crystal. One good example is doping LiNbO$_3$ with Fe ions, which enhances photorefractive effect and makes it a perfect medium for holographic storage$^{[1]}$. Another example is to use MgO as a dopant, which is a counter-measurement for the photorefractive damage and can greatly improve its high power performance. MgO doped LiNbO$_3$ is now widely used in nonlinear optical devices$^{[2]}$, because under these circumstances high optical damage threshold is often required. Albeit, stoichiometric LiNbO$_3$ can still be created if special care is taken during the growing process, such as using the double crucible$^{[3]}$ or vapor transport equilibration techniques$^{[4]}$.

Because of the ferroelectric nature of LiNbO$_3$ crystal, it can be domain-engineered. By applying high voltage electrical pulse against its z axis, the ferro domains through the crystal can be reversed$^{[5]}$. The periodically polled LiNbO$_3$ (PPLN) is a crucial material for quasi-phase matched nonlinear optical applications such as second harmonic generation (SHG), difference frequency generation (DFG), and
optical parametric oscillation (OPO). Taking advantage of its largest nonlinear tensor $d_{33}$, PPLN is able to deliver impressive efficiency much higher than other crystals such as periodically polled KTiPO$_4$ (PPKTP). Through domain engineering, nanostructures can also be created on the crystal surface$^{[6]}$, which have generated many interesting topics and offered another path of tailoring the crystal property.

![Diagram of PPLN crystal axis and high voltage poling](image)

**Fig. 1.1.** (a) The preparation method of PPLN. (b) Photograph of PPLN wafers.

*(Picture credit: Crystal Tech., Inc.)*

1-2. LiNbO$_3$ Optical Waveguides

Over the past decades, intense research interests have been placed on fabricating high quality optical waveguides on LiNbO$_3$, because of its good prospection in many applications such as high-speed modulators and nonlinear wavelength convertors. The most popular way of fabricating optical waveguide on LiNbO$_3$ substrate is through ion doping method such as Ti in-diffusion or annealed proton-exchange (APE)$^{[7][8]}$. After doping, the refractive index increases in the doped area, and thus forms waveguiding channels. In recent years, some other advanced techniques have merged, for example, the etched waveguide and precise machined ridge waveguide$^{[9][10]}$. The details of various fabrication techniques will be discussed in the following chapters.
Nowadays, although the fabrication techniques on semiconductor optical waveguide are quite mature, and many of them have already become industrial standard process, high quality LiNbO$_3$ waveguide is much more difficult to fabricate, and only a few of them have practical value. Traditional APE waveguides have poor light confinement, and wet etching method is not compatible with periodically polled structure. Dry etched and precisely machined ridge type waveguides have good performances, but the fabrication process is tedious and needs improvements. Exploring new fabrication method and improving the performance of LiNbO$_3$ device has become a lift-time professional pursuit for many researchers and engineers.

1.3. Purpose of This Study

The main purpose for this study is to sum up the existing LiNbO$_3$ waveguide fabrication techniques as well as explore new methods of fabricating high quality LiNbO$_3$ ridge waveguides compatible with periodically polled substrates. Our final goal is to fabricate a wavelength conversion device with remarkable performance based on PPLN ridge waveguide.

In the following chapters, a novel LiNbO$_3$ ridge waveguide fabrication method by the combination of annealed proton-exchange and precise diamond blade dicing techniques will be fully illustrated with both theoretical analysis and experimental results. The experimental results considering the waveguide quality will be systematically compared and discussed. Based on this research, green light SHG experiment is successfully conducted using MgO doped PPLN APE ridge waveguide. The acquired data has shown much higher conversion efficiency than those achieved by traditional ion-doped channel waveguides. This research will provide a new platform for various LiNbO$_3$ waveguide devices such as nonlinear interaction or high-speed modulation.
Chapter 2. History of LiNbO$_3$ Waveguide

2-1. Classification of LiNbO$_3$ Waveguide

In the year 1974, the first plausible fabrication technique for LiNbO$_3$ waveguide was proposed by R. V. Schmidt and I. P. Kaminow at Bell Laboratory$^7$. Since then, more and more researchers began to focus on this topic, and various methods have been reported. The existing LiNbO$_3$ waveguides can be classified into three kinds: ion-doped slab/channel waveguide, ion-doped ridge waveguide and wafer bonded slab/ridge waveguide. Each kind represents a typical fabrication sequence and generates waveguides with similar characteristics. The major literature contributions are listed as follows:

(I) Ion-doped slab/channel waveguide
   Metal in-diffusion (Ti, Ni, Zn, etc.)$^7$$^{11}$$^{12}$
   Proton-exchange / annealed proton-exchange$^8$$^{13}$
   Ion implantation$^{14}$$^{15}$

(II) Ion-doped ridge waveguide
   Ion-doping + (-z) axis wet etching$^9$$^{16}$$^{17}$$^{18}$$^{19}$$^{20}$
   Ion-doping + RIE$^{21}$$^{22}$$^{23}$

(III) Wafer bonded slab/ridge waveguide
   Adhered wafer bonding + wafer thinning + etching$^{24}$
   Direct wafer bonding + wafer thinning + etching$^{25}$$^{26}$
   Adhered wafer bonding + wafer lapping + diamond blade dicing$^{10}$$^{27}$$^{28}$$^{29}$
   Direct wafer bonding + wafer lapping + diamond blade dicing$^{30}$$^{31}$$^{32}$$^{33}$$^{34}$

2-2. Fabrication Techniques

As above-mentioned, the three different types of LiNbO$_3$ waveguide have their particular fabrication processes, which distinguish them from each other. The major fabrication techniques will be discussed in detail below.
2-2-1. Ion Diffusing/Exchanging

In the 1970s, researchers found that by diffusing metal ions into LiNbO₃ under high temperature, the refractive index of the surface layer could be slightly increased. Although the index rises only in an order of $10^{-3}$, it is sufficient to form waveguide channels.

The most famous method is Ti in-diffusion. Using the matured photolithography and metallization techniques, waveguides can be well defined on LiNbO₃ wafer surface. After several hours of thermal diffusion at around 1000°C, a Gaussian-like index profile is generated with a depth of several micrometers. This kind of waveguide is guiding to both TM and TE mode, and can be formed on whether X, Y or Z cut LiNbO₃ crystal. Although with an asymmetric beam profile, the waveguides can be made into single mode, and the propagation loss is as low as $0.05 \text{ dB/cm}$\textsuperscript{16}. However, later research work has found out that the Ti diffused waveguide suffers from severe photorefractive damage, which undermines its performance when operated under high power density.

Fig. 2.1. A typical process of Ti in-diffusion, which mainly contains four steps: Ti deposition, waveguide definition via photolithography, waveguide pattern etching out and substrate cleaning, and finally thermal in-diffusion.
Another popular way of making ion-diffused waveguide is by proton-exchange (PE). Li⁺ ions in LiNbO₃ can be easily replaced by H⁺ ions in hot acid, and transform into HNbO₃. After reaction, a thin hydrogen-rich layer, usually several tens to hundreds of nanometers, can be formed on the crystal surface. If the reaction takes place in strong acid such as HNO₃ or H₂SO₄, the exchange is complete, and the crystal structure will be destroyed by this chemical reaction. However, if a weak acid is used, such as benzoic acid, the Li⁺ ions will be partially replaced by H⁺ ions and forms HₓLi₁₋ₓNbO₃ hybrid crystal. If the reaction condition is properly selected, the crystal structure will largely remain untouched.

![Fig. 2.2. H⁺ concentration profile of a z-cut LiNbO₃ with 35min PE time at 225°C. Measurement is carried out using secondary ion mass spectroscopy (SIMS).](Picture credit Sandeep T. Vohra et al.[35]).

Using this method, proton-exchanged waveguide can be created on LiNbO₃ wafer surface, because the exchanged area will have a comparatively large refractive index increase for extraordinary light, usually around 0.1[8]. This kind of waveguide features with single polarization and better light confinement than Ti in-diffused waveguide, but at the same time, it has high propagation loss and degenerated nonlinear
Further study indicates that by annealing the proton-exchanged crystal at around 300-400°C for a certain time, the $H^+$ ions will diffuse further into the crystal, and the original crystal property of ion exchanged area can thus be restored. As a result, high quality waveguide with low propagation loss, single polarization, and almost the same nonlinear coefficient as bulk material can be created.

![Diagram of LiNbO₃ processing steps](image)

**Fig. 2.3.** LiNbO₃ benzoic acid proton-exchange and annealing processes. First, an aluminum film is deposited on the wafer as a mask; then waveguide pattern is defined using photolithography and etching; proton-exchange is carried out in melted benzoic acid at around 200°C; at last, the crystal is annealed at 300-400°C.

There are also some other methods to produce ion doped LiNbO₃ waveguide. For example, using Zn or Ni in-diffusion, or a combination of Ti in-diffusion and proton-exchange. Ion implantation is also a viable way to make waveguides. By implanting $H^+$, $He^+$ or $O^+$ ions into the crystal, a low refractive index optical barrier is formed, which makes the light conducting in the unmodified surface region. The characteristics of such waveguides can be controlled by changing the implantation energy and dose.
To sum up, ion doping is a matured and widely used method to produce low loss LiNbO₃ waveguide. The fabrication process is easy to carry out, and results are repeatable. Moreover, most of the techniques are compatible with periodically polled substrate. However, light confinement in ion doped channel waveguide is weak and asymmetric, and some of the dopant such as Ti causes severe photorefractive damage, which hampers the waveguide performance under high power operation.

2-2-2. LiNbO₃ Wet Etching

Etching techniques have been widely used in making LiNbO₃ ridge structures, and through the combination of etching and ion doping or wafer bonding, ridge waveguide can be fabricated, which provides much better light confinement than traditional ion doped channel waveguides. However, although etching of semiconductor materials is quite mature, etching of LiNbO₃ has caused a difficult problem because of its unique chemical properties and crystal structures.

HF solution is used to carry out LiNbO₃ wet etching. From a chemical point of view, HF and LiNbO₃ have the following reactions:

\[
\text{LiNbO}_3 + 9\text{HF} = \text{LiHF}_2 + \text{H}_2\text{NbF}_7 + 3\text{H}_2\text{O}
\]  

(2-1)

However, the experimental results have demonstrated different characteristics along different crystal axis. The etching rate along \(-z\)-axis is the fastest one, while \(+z\) direction is almost resistant to the reaction. Reactions along \(x\) and \(y\)-axis are also different from each other, but they are all much slower than the \(-z\) direction. As a result, ridge structure can be easily created on the \(-z\) surface of LiNbO₃ wafer, while it is difficult to carry out from the other directions. In order to explain this phenomenon, we must know what indeed happened during this reaction.
Fig. 2.4. LiNbO$_3$ ridge waveguide fabricated by wet etching on $-z$ surface. HF and HNO$_3$ mixed acid is used to carry out the etching. *(Photo credit H. Hu et al.\textsuperscript{[9]}\textsuperscript{[16]})*

The reaction dynamics is believed to be a two step procedure\textsuperscript{[36]}. First, the crystal surface, which is covered with oxygen ions, is protonized by hydrogen ions in the acidic solution, and forms water molecule; then fluorine ions are attached on Nb ions, and pull it away from the crystal lattice. After that, the protonization happens again in a cycled manner, and Li ions just dissolve into the solution. Because LiNbO$_3$ is a ferroelectric crystal, it has positively charged electric field on the $+z$ surface. As a result, it forms a barrier for protonization process and blocks the reaction. In a contrary, reactions on $-z$ surface are accelerated by the negative charges. Since the intrinsic electrical polarization is very strong, the result demonstrates distinguishing etching rate on the two different surfaces.

The etching speed on $-z$ surface largely depends on temperature and the composition of the etchant. At higher temperature, the reaction takes place much faster, but high temperature is definitely not welcome because it will generate toxic HF fumes. For the HF solution, the highest concentration does not provide the highest reaction rate, because of the decreased H$^+$ ion concentration in saturated HF acid, which is caused by the formation of HF molecule chains. In order to fortify H$^+$ ion concentration, HNO$_3$ is often added as an ingredient.
Fig. 2.5. Wet etching rate under different HF and HNO₃ mixed acid volume ratio.

Fig. 2.6. Surface morphology of LiNbO₃ ridge structure etched on −z surface using HF/HNO₃ mixed acid with different ratio. From (a)-(d), HF:HNO₃ volume ratio is 100%, 90%, 80%, and 70% respectively.
In our study, we have found out that a mixture of 49% HF and 68% HNO₃ with a volume ratio 8:2 gives the highest etching rate. However, the etched surface roughness begins to increase drastically when HF volume ratio exceeds 70% (Fig.2.6), as a result, the best volume ratio of HF and HNO₃ acid for LiNbO₃ wet etching should be 7:3.

In order to create ridge waveguide on LiNbO₃ +z surface by wet etching, another method is proposed. Researchers found out that the after proton-exchange the exchanged area can be etched by HF acid because the ferroelectric field is partially destroyed during exchanging. Using this method, ridge waveguide can be created on +z surface. However, because the surface roughness of etched proton-exchanged area is considerably high, the propagation loss of such waveguide is also higher than that fabricated on -z surface.

Wet etching method is ideal to produce LiNbO₃ waveguides in large area with low propagation loss and good uniformity. However, the major drawback is its non-compatibility with periodically polled structure.

2-2-3. LiNbO₃ Dry Etching

As plasma technology advances, dry etching method has also been introduced in LiNbO₃ ridge structures fabrication. There are basically two principles. The first one is physical heavy ion bombardment using Ar gas; the second one is reactive etching, using fluoride gases such as CF₄ or SF₆. Fluorine ions react with LiNbO₃ and thus the etching rate is much higher. However, because LiF is a non-volatile compound, it will re-deposit on the substrate and forms a barrier for further reaction. As a result, the etching rate decreases fast as reaction goes on, and the surface roughness of etched area is not satisfying.

To improve this situation, a combination of heavy ion milling and reactive etching is often used. Fluoride gas helps etching the LiNbO₃ substrate, while heavy ions help cleaning the re-deposited LiF. Inductively coupled plasma (ICP) or magnetic neutral
loop discharge (NLD) source is often used to carry out the etching more effectively. To further improve the etching quality, another method is taking advantage of the PE process. After PE process, the Li ions are substituted by H ions, which could alleviate the problem of LiF re-deposition during etching.

Fig. 2.7. Ridge structure fabricated on z-cut LiNbO$_3$ substrate using Ar/C$_3$F$_8$ NLD plasma dry etching. (Photo credit W. S. Yang et al.[22])

![Fig. 2.7](image)

(a) (b)

Fig. 2.8. LiNbO$_3$ photonic crystal structure etched out using proton-exchange assisted dry etching. CHF$_3$/Ar ICP plasma is used; the structure is created on x-cut crystal. (Photo credit H. Hu et al.[23])

![Fig. 2.8](image)

(a) (b)

The proton-exchange depth is usually very shallow, typically on the order of 1 micron, deeper than that, the advantage of such proton-exchange assisted method is
lost. Except this, the surface roughness of the etched area remains a problem, just like the scene in wet etching. Albeit, dry etching technique is still a plausible way for mass production of LiNbO\textsubscript{3} ridge waveguide. As the technology advances, it is hopeful that better solutions can be found in the near future.

2-2-4. Wafer Bonding

In order to produce LiNbO\textsubscript{3} waveguide without ion doping, wafer bonding techniques are introduced as a way for thin film formation. After plasma treatment, LiNbO\textsubscript{3} crystal wafer can be bonded to another foreign substrate, and then using lapping or some other techniques, the crystal is thinned down to several microns and thus forms a thin film, after that ridge structures can be created using etching or precise dicing.

There are many reported cases that LiNbO\textsubscript{3} wafer is successfully bonded to other substrates (Ref. [24]-[34]); typically can be sorted into two kinds. The first class is LiNbO\textsubscript{3} bonded to another crystal using interfacial mediums such as optical epoxy; the second class is direct bonding between two crystals with plasma treatment.

LiNbO\textsubscript{3} crystal can be easily bonded to other substrates after spin-coated with a thin layer of optical epoxy. Theoretically speaking, LiNbO\textsubscript{3} wafer can be bonded to any other substrate using this method, no matter of its chemical property. Whatever it sounds so easy, in practical, it is totally a different thing. Very high precision machinery is needed to carry out the bonding. Thickness variations across the whole sample must be controlled at a nanometer scale, in order to avoid large thickness fluctuation of the thin film.
Another bonding scheme is utilizing plasma surface activation. After plasma treatment, the bonding interfaces are covered by high-energy radicals and dangling bonds. After contact, the active molecules react with each other and form permanent chemical bonds.

According to the nature of bonded substrate, LiNbO$_3$ direct bonding can be divided into two kinds. The first one is bonding LiNbO$_3$ with ionic crystal, such as LiTaO$_3$. As described in literatures, after O$_2$ or Ar plasma activation, LiNbO$_3$ and LiTaO$_3$ crystals are aligned and bonded together in ultra high vacuum chamber. The plasma activation parameters are crucial to success bonding, if the plasma activation is too gentle, insufficient chemical bonds are generated. If the activation is too heavy, the bonding interface may be damaged under intense ion bombardment. In order to fortify the chemical bonding between two interfaces, annealing is often carried out as an after process.
Fig. 2.10. LiNbO$_3$ ridge waveguide fabricated using crystal bonding, ion slicing and Ar ion milling. Waveguiding region contains three layers: LiNbO$_3$ thin film, SiO$_2$ insulating layer, and LiNbO$_3$ substrate. (Photo credit H. Hu et al.$^{[19]}$)

The second kind of direct bonding is between LiNbO$_3$ and covalent materials such like semiconductors. In addition, to avoid serious light absorption in visible wavelength, an optical barrier layer such as SiO$_2$ thin film is inserted between the two interfaces. However, because of the tremendous differences in thermal expansion coefficients of the two substrates’ properties, thermal annealing process is almost impossible in this case. Several tens of degrees change in temperature will cause fracture or de-bonding. As a result, the bonding strength is often very low. Until now, there is no successful application device built on such a hybrid structure yet.

2-2-5. Wafer Ion Slicing

After wafer bonding, the bonded LiNbO$_3$ must be thinned down to microns to form a waveguiding thin film. As worth mentioning, one way to realize it is crystal ion slicing.

As described in Ref. [25], the general process is: A certain dose of He ions are implanted into LiNbO$_3$, the implanted depth can be controlled by varying the ion energy, it can vary from tens of nanometers to several microns. Then after heating it up to around 200ºC, the thin film will be sliced away from bulk substrate. Chemical mechanical polishing is often applied to the sliced interface to restore smoothness and
reduce light scattering loss. Using this method, a very thin LiNbO₃ film can be fabricated and integrated on another substrate. There are successful cases of device fabrication reported [25][26], as showed in Fig. 2.10, based on LiNbO₃-SiO₂-LiNbO₃ ridge waveguide structure. It is a plausible way to fabricate large area thin films with good uniformity.

![Flow chart of LiNbO₃ ion slicing process](image)

Fig. 2.11. Flow chart of LiNbO₃ ion slicing process, which mainly contains four steps: thin film definition by ion implantation, PECVD, wafer bonding and slicing.

2-2-6. Precise Machining

The fabrication method of LiNbO₃ ridge waveguide using precise machining was first introduced by several Japanese companies. After bonding LiNbO₃ wafer to a foreign substrate, the wafer is first grinded and then chemical mechanical polished to form a thin film with good smoothness and uniformity. Then, ridge waveguide is cut out by high precision diamond blade dicing. All processes are accomplished by highly precise mechanical instruments [30]-[34].

Apparently, to control the film thickness and ridge waveguide dimensions with accuracy below one micrometer is a challenging task. Fortunately, the advancing technologies in modern mechanical engineering have offered us good solutions. The
grinding wheel and dicing blade are made of synthetic diamond particles embedded in holding matrixes such as metal or resin medium. The exposing diamond particles provide effective cutting with the rotation of the spindle. The size and concentration of diamond particles can be carefully controlled in several microns, which have enabled precise controlling of the microstructures.

![SEM image of the tip of a dicing blade](Picture credit DISCO Corp.)

Fig. 2.12. SEM image of the tip of a dicing blade, showing the embedded diamond particles in holding matrix. (Picture credit DISCO Corp.)

Ridge waveguides fabricated by precise dicing have demonstrated excellent characteristics, as reported in recent years\textsuperscript{30}-\textsuperscript{34}. It is a reliable method to produce LiNbO\textsubscript{3} waveguides with low propagation loss and tight light confinement. Moreover, the nonlinear property of LiNbO\textsubscript{3} layer remains the same as bulk material. The only problem is that the techniques require highly precise mechanical instruments.

2-3. Selected Applications of LiNbO\textsubscript{3} Ridge Waveguide

2-3-1. Optical Modulation

As the advancement of information technology grows more and more rapidly, the requirement on large capacity optical transmission network keeps increasing faster than any period in history. External optical modulator is one of the key components for high speed fiber communications, and it has been widely studied over the past decades\textsuperscript{37,38}. LiNbO\textsubscript{3} is of great interest in this field of application, for its excellent
performance in extinction ratio, insertion loss and optical transparency. Single channel modulation with a bandwidth of over 40 GHz has already been commercialized, and even higher bandwidth can be achieved using multiplexing technologies.

In order to further increase the modulation speed as well as shrink device volume and save operation costs, lower driving voltage is required on the electrical drivers. LiNbO$_3$ ridge waveguide is an excellent candidate for higher bit rate modulators, because the much better light field confinement has enhanced the interaction between light and microwave. Further studies have suggested that ridge structure is also helpful in impedance matching between electrical and optical components. Although the fabrication process for ridge type LiNbO$_3$ waveguide is still under developing, it is hopeful that such ridge type modulators can be industrialized in the near future.

![Fig. 2.13. LiNbO$_3$ Mach-Zehnder modulator built on (a) conventional channel waveguide, and (b) ridge waveguide. (Picture credit D. Janner et al.)](image)

2-3-2. Nonlinear Wavelength Conversion

For solid state lasers, since active medium for most frequencies in the visible electromagnetic spectrum is unavailable, wavelength conversion component must be used in order to create lasers at these wavelengths. Miniaturized visible lasers have caught special interests because of their potential markets in advanced display technologies. While RGB light sources are basic components for color displays, semiconductor lasers can only provide solutions at red and blue light region. Green
light generation using GaN based compounds still remains a painful work, because material with direct bandgap for 532 nm wavelength is hard to grow.

In addition, at infrared region, 775 nm light generation using 1550 nm light SHG is useful in optical communication. In addition, mid-infrared radiation around 2-5 μm wavelength, which can be generated using infrared DFG, is highly desirable for environmental gas composition monitoring.

PPLN is a domain-engineered crystal for quasi phase matching nonlinear interaction, which takes advantage of its ferroelectric property and largest $d_{33}$ nonlinear coefficient. Bulk PPLN is often used as an intra-cavity wavelength conversion component, for example in SHG, DFG or OPO lasers. A single pass SHG and DFG laser using optical waveguide is another plausible solution. Since the nonlinear conversion efficiency is closely related to light power density, in order to acquire high conversion efficiency in LiNbO$_3$ waveguide, ridge structure is more attractive than traditional channel waveguide, and single pass conversion efficiency can achieve considerably high value. The fabrication techniques described in earlier paragraphs are not all suitable to create waveguide on PPLN. For example, the wet chemical etching method is highly selective along z directions, which will generate grating structures rather than a smooth waveguide on PPLN chips. In addition, to avoid photorefractive damage under high light power intensity, MgO doped LiNbO$_3$ crystal is used instead of congruent crystal.

Successful wavelength conversion experiment using LiNbO$_3$ ridge waveguide have been demonstrated in recent years’ publications, showing much better improvements than any records ever in history. 775 nm light SHG with normalized conversion efficiency of 370%/W·cm$^2$ has been demonstrated by Sunao Kurimura et al. at National Institute for Materials Science, Japan, using a QPM adhered x-cut Zn-doped LiNbO$_3$ ridge waveguide$^{29}$. Green light generation with the highest conversion efficiency of 72.9% has been achieved by Hong Ky Nguyen et al. at
Corning Inc., using bonded MgO:LiNbO$_3$ ridge waveguide and a high power DBR laser\cite{30}. Another example for wavelength up-conversion is blue light generation with peak conversion efficiency of 58% was reported by M. Iwai et al. at NGK Insulators Ltd., using an adhesive bonded MgO:LiNbO$_3$ ridge waveguide\cite{28}. 3 $\mu$m mid-infrared light generation with 40%/$\text{W}$ efficiency was also demonstrated by Osamu Tadanaga et al. at NTT Corporation, using infrared DFG in a 50 mm long direct bonded QPM-LN ridge waveguide\cite{32}.

Fig. 2.14. Fabrication process of adhesive bonded PPLN ridge waveguide, containing three major steps: adhesive bonding, wafer lapping and polishing, and precise diamond blade dicing.
Chapter 3. Theory of Second-Order Nonlinear Interaction

3-1. Nonlinear Susceptibility

When an electromagnetic wave interacts with matter, polarization occurred inside the medium because the electrons get affected by external electric force. The polarization response can be expressed using Fourier series expansion:

\[ P = \varepsilon_0 \left[ \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots \right] \]  \hspace{1cm} (3-1)

The first term \( P = \varepsilon_0 \chi^{(1)} E \) describes the linear response of the material, and the following term \( P = \varepsilon_0 \chi^{(2)} E^2 \) describes second-order nonlinear polarization, where \( \chi^{(n)} \) is the n-th order susceptibility of the medium. In anisotropic medium, \( \chi \) is no longer a scalar but a tensor that links vectors \( P \) and \( E \). To be more specifically, when two frequencies \( \omega_1 \) and \( \omega_2 \) are present, the second order polarization can be written as:

\[
\frac{1}{\varepsilon_0} P(\omega_1, \omega_2) = \sum_{jk} \chi_{ijk} (\omega_1, \omega_2) E_j (\omega_1) E_k (\omega_2) \]  \hspace{1cm} (3-2)

The nonlinear polarization will emit new frequencies. For example, in the case of second harmonic generation, we can see \( P(\omega) \propto E^2 (\omega) \), and if we return to the time domain and denote \( E(t) = E_0 \cos(\omega t) \), then we have:

\[
P(\omega) \propto E_0^2 \cos^2(\omega t) = \frac{E_0^2}{2} [1 + \cos(2\omega t)] \]  \hspace{1cm} (3-3)

It contains an optical rectified signal and a new doubled frequency. Usually, the \( \chi^{(2)} \) effect is 7~8 orders of magnitude smaller than \( \chi^{(1)} \), and so does the higher order nonlinear susceptibility. Only when the external electric field strength is on the order of the characteristic atomic electric field strength (i.e. in hydrogen atom), the second order nonlinear effect becomes comparable to the linear effect.
3-2. Maxwell Equation for Nonlinear Interaction

We shall start from Maxwell equations and derive a suitable way for SHG calculation. As we know, when light travels in dielectric medium, the Maxwell equations have the following expression:

\begin{align*}
\nabla \times \mathbf{E} &= -\frac{\partial \mathbf{B}}{\partial t} \\
\nabla \times \mathbf{H} &= \mathbf{j} + \frac{\partial \mathbf{D}}{\partial t} \\
\nabla \cdot \mathbf{D} &= \rho \\
\nabla \cdot \mathbf{B} &= 0
\end{align*}

where the free current and density electric displacement are \( \mathbf{j} = \sigma \mathbf{E} \) and \( \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \).

The polarization induced by external electric field can be separated in linear and nonlinear part. We denote the nonlinear part as \( \mathbf{P}_{\text{NL}} \), and then the total polarization can be expressed as:

\[ \mathbf{P} = \varepsilon_0 \chi \mathbf{E} + \mathbf{P}_{\text{NL}} \]  

(3-8)

Noticing that \( \varepsilon = \varepsilon_0 (1 + \chi) \), then eq. 3-5 becomes:

\[ \nabla \times \mathbf{H} = \sigma \mathbf{E} + \varepsilon \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_{\text{NL}}}{\partial t} \]  

(3-9)

From the Eq. 3-4, taking curl of both sides:

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

(3-10)

By combining the up two equations, we get:

\[ \nabla \times \nabla \times \mathbf{E} = -\mu \frac{\partial}{\partial t} \left( \sigma \mathbf{E} + \varepsilon \frac{\partial \mathbf{E}}{\partial t} + \frac{\partial \mathbf{P}_{\text{NL}}}{\partial t} \right) \]  

(3-11)
Using the relations:

\[ \nabla \times \nabla \times \mathbf{E} = \nabla (\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} \quad (3-12) \]

By noticing \( \nabla \cdot \mathbf{E} = 0 \) in dielectric medium, the equation becomes:

\[ \nabla^2 \mathbf{E} = \mu \frac{\partial}{\partial t} \left( \sigma \mathbf{E} + \epsilon \frac{\partial}{\partial t} \mathbf{E} + \frac{\partial}{\partial t} \mathbf{P}_{nl} \right) \]

\[ \nabla^2 \mathbf{E} - \mu \sigma \frac{\partial}{\partial t} \mathbf{E} - \mu \epsilon \frac{\partial^2}{\partial t^2} \mathbf{E} = \mu \frac{\partial^2}{\partial t^2} \mathbf{P}_{nl} \quad (3-13) \]

3.3. Coupled Wave Equations

Here we consider second order three wave interactions. Suppose we have two incoming plane wave of frequencies \( \omega_1 \) and \( \omega_2 \), propagating along z-axis with linear polarization, the electrical fields can be expressed as follows:

\[ E_1(z,t) = E_1(z) \exp(ik_1z - i\omega_1t) \quad (3-14) \]

\[ E_2(z,t) = E_2(z) \exp(ik_2z - i\omega_2t) \quad (3-15) \]

We denote the one of the new generated frequency \( \omega_1 + \omega_2 \) as \( \omega_3 \), and then its electric field and induced second order nonlinear polarization can be written as:

\[ E_3(z,t) = E_3(z) \exp(ik_3z - i\omega_3t) \quad (3-16) \]

\[ P_{nl}(z,t) = \varepsilon \chi_{\text{eff}} E_1(z)E_2(z) \exp[i(k_1 + k_2)z - i(\omega_1 + \omega_2)t] \quad (3-17) \]

where \( \chi_{\text{eff}} \) is the effective nonlinear susceptibility.

By substituting \( E_3 \) into the deduced wave equation eq. 3-13, the left side becomes:

\[ \frac{\partial^2}{\partial z^2} E_3(z,t) - \mu \sigma \frac{\partial}{\partial t} \mathbf{E} - \mu \epsilon \frac{\partial^2}{\partial t^2} \mathbf{E} \]
Assuming that the change of wave package form over a certain distance is very small, which is also know as the “slowly varying amplitude approximation”, we take the second order spatial derivative to be zero:

\[
\frac{d^2}{dz^2} E_3(z) = 0 \tag{3-19}
\]

Noticing that \(\mu_0 \omega^2 - k^2 = 0\), there will be only two terms left:

\[
\text{Eq.Left} = 2ik_3 \exp(ik_3z - i\omega_3t) \frac{d}{dz} E_3(z) + i\omega_3 \mu_0 E_3(z) \exp(ik_3z - i\omega_3t) \tag{3-20}
\]

Then the right side of wave equation is written as:

\[
\text{Eq.Right} = \mu \frac{d^2}{dt^2} P_{nl} = -\mu_0 \varepsilon_0 (\omega_1 + \omega_2)^2 \chi_{\text{eff}} E_1(z) E_2(z) \exp[i(k_1 + k_2 - k_3)z - i(\omega_1 + \omega_2)t]
\]

Equating the two sides, so we get:

\[
2k_3 \frac{d}{dz} E_3(z) = i\mu_0 \varepsilon_0 \omega_3^2 \chi_{\text{eff}} E_1(z) E_2(z) \exp[i(k_1 + k_2 - k_3)z] - \omega_3 \mu_0 E_3(z) \tag{3-22}
\]

In most cases of dielectric optical material, we have \(\mu = \mu_0\); and if we assume the medium has no absorption loss to the incident light, we have \(\sigma = 0\), then the equation can be simplified as:
\[ \frac{d}{dz} E_{1}(z) = \frac{i\omega_{1}}{n_{1}c_{0}} d_{\text{eff}} E_{2}^{*}(z) E_{3}(z) \exp(i\Delta k z) \] (3-23)

In this equation, we denote \( d_{\text{eff}} = \frac{\chi_{\text{eff}}}{2} \) as the effective nonlinear coefficient, and \( \Delta k = k_{1} + k_{2} - k_{3} \), which is the wave vector mismatch. Note that the energy flow during wave mixing is not unidirectional. Following the same manner, we get the other two equations for \( E_{1} \) and \( E_{2} \) (* means conjugate):

\[ \frac{d}{dz} E_{1}(z) = \frac{i\omega_{2}}{n_{2}c_{0}} d_{\text{eff}} E_{1}^{*}(z) E_{3}(z) \exp(-i\Delta k z) \] (3-24)

\[ \frac{d}{dz} E_{2}(z) = \frac{i\omega_{3}}{n_{3}c_{0}} d_{\text{eff}} E_{3}^{*}(z) E_{2}(z) \exp(-i\Delta k z) \] (3-25)

### 3-4. Second Harmonic Generation

#### 3-4-1. Non-depletion Approximation

Under the condition \( \omega_{1} = \omega_{2} = \omega \), we can see that the three wave coupling equation could be reduced into two; the original electric field \( E_{1} \) and \( E_{2} \) merges into one large field \( E_{\omega} \), and its amplitude equals the sum of two separate ones \( (E_{1}^{2} + E_{2}^{2} = E_{\omega}^{2}/2) \). Therefore, we get the coupled wave equations for SHG:

\[ \frac{d}{dz} E_{\omega}(z) = \frac{i\omega}{n_{\omega}c_{0}} d_{\text{eff}} E_{\omega}^{*}(z) E_{2\omega} \exp(-i\Delta k z) \] (3-26)

\[ \frac{d}{dz} E_{2\omega}(z) = \frac{i\omega}{n_{2\omega}c_{0}} d_{\text{eff}} E_{\omega}^{2}(z) \exp(i\Delta k z) \] (3-27)

Under non-depletion assumption, which means the strength of \( E_{\omega} \) does not change during the entire interaction, we can integrate the equation from 0 to a length of \( L \) as:
After calculation and several steps of transformation, \( E_{2\omega} \) becomes:

\[
E_{2\omega}(z) = i\omega L d_{\text{eff}} E_{\omega}^2 \int_{0}^{t} \exp(i\Delta k z) dz = \frac{\omega}{n_{2\omega} c_0} d_{\text{eff}} E_{\omega}^2 \left( \frac{\Delta k L}{2} \right) \exp(i\Delta k L/2) \quad (3-29)
\]

The intensity of an optical field can be expressed as:

\[
I = \frac{1}{2} n c_0 E_0 E^* \quad (3-30)
\]

So the SHG intensity \( I_{2\omega} \) can be expressed as a function of fundamental intensity \( I_\omega \):

\[
I_{2\omega} = \frac{2\omega^2 L^2 d_{\text{eff}}^2 I_{\omega}^2 \sin^2 \left( \frac{\Delta k L}{2} \right)}{n_{2\omega}^2 n_\omega^2 c_3 e_0} = I_{\text{MAX}} \sin^2 \left( \frac{\Delta k L}{2} \right) \quad (3-31)
\]

Hence, the conversion efficiency is:

\[
\eta_{\text{SHG}} = \frac{I_{2\omega}}{I_\omega} = \frac{2\omega^2 L^2 d_{\text{eff}}^2 I_{\omega} \sin^2 \left( \frac{\Delta k L}{2} \right)}{n_{2\omega} n_\omega c_3 e_0} = \eta_{\text{MAX}} \sin^2 \left( \frac{\Delta k L}{2} \right) \quad (3-32)
\]

Fig. 3-1. Relationship between phase mismatching factor \( \Delta k L \) and SHG conversion efficiency \( \eta_{\text{SHG}} \). \( \eta_{\text{MAX}} \) is defined as in Eq.(3-32).
From this equation, we can obtain some important information about SHG. First, the conversion efficiency is proportional to the fundamental power, but the SHG power is proportional to the square of input power. Second, the SHG process is proportional to the square of nonlinear coefficient. Third, the SHG process is proportional to the square of $L$ times a $\text{sinc}$ function, which involves the phase mismatching vector $\Delta k$. This means that the SHG power will not keep increasing as the wave propagates; when the phase-matching condition is running out, the SHG is stopped. Thus, practically certain treatment is needed to keep the two wave fronts in-phase.

3-4-2. SHG with Depletion

When the SHG signal grows to a remarkable level as the fundamental power, non-depletion model is not effective anymore. The energy back flow from $E_{2\omega}$ to $E_{\omega}$ must be considered. That means the $E_{2\omega}$ cannot be integrated separately anymore, the two coupled wave equations must be solved simultaneously.

To simplify our discussion and focus on the light power issue, we consider only the wave amplitude under total phase matching. We set $\Delta k=0$, and $n_{2\omega}=n_{\omega}=n$, the wave equation becomes:

\[
\frac{d}{dz} A_{2\omega}(z) = \frac{\omega}{nc_0} d_{\text{eff}} A_{\omega}^2(z) \tag{3-33}
\]

\[
\frac{d}{dz} A_{\omega}(z) = \frac{\omega}{nc_0} d_{\text{eff}} A_{\omega}(z) A_{2\omega}(z) \tag{3-34}
\]

From the conservation of energy, we have $A_{\omega}^2 + A_{2\omega}^2 = A_{\omega}^2(0) = \text{Const.}$, and by substituting it into the equation, we get:

\[
\frac{d}{dz} A_{2\omega}(z) = \frac{\omega d_{\text{eff}}}{nc_0} [A_{\omega}^2(0) - A_{2\omega}^2(z)]
\]

- 27 -
Using the following calculus relation:

$$\int \frac{dx}{a^2 - x^2} = \frac{1}{a} \tanh^{-1} \frac{x}{a}$$  \hspace{1cm} (3-36)

We can get the analytical solution for $A_{2\omega}$ as:

$$A_{2\omega}(z) = A_{\omega}(0) \tanh \left[ \frac{\omega_d\ell}{n_c} z A_{\omega}(0) \right]$$  \hspace{1cm} (3-37)

So the SHG light intensity $I_{2\omega}$ and fundamental light intensity $I_{\omega}$ is:

$$I_{2\omega}(z) = I_{\omega}(0) \tanh^2 \left[ \frac{\omega_d\ell}{n_c} z \sqrt{\frac{2I_{\omega}(0)}{n_c\varepsilon_0}} \right]$$  \hspace{1cm} (3-38)

$$I_{\omega}(z) = I_{\omega}(0) - I_{2\omega}(z) = I_{\omega}(0) \text{sech}^2 \left[ \frac{\omega_d\ell}{n_c} z \sqrt{\frac{2I_{\omega}(0)}{n_c\varepsilon_0}} \right]$$  \hspace{1cm} (3-39)

![Fig. 3.2. SHG interaction under depletion and perfect phase matching. Parameter $L_{\text{SHG}} = \frac{n_c}{\omega_d \ell} \sqrt{\frac{n_c\varepsilon_0}{2I_{\omega}(0)}}$, corresponding with Eq.(3-38) and Eq.(3-39).](image)
3-5. Quasi Phase Matching

We have already derived the SHG power change under non-depletion condition. Now we examine it again by setting $z$ as the variable rather than $\Delta k$:

$$I_{2\omega}(z) = \frac{2\omega^2 z^2 d_{\text{eff}}^2 I_\omega^2 \sin^2 \left( \frac{\Delta k z}{2} \right)}{n_{2\omega}^2 n_\omega^2 c^3 \varepsilon_0} = \frac{8\omega^2 d_{\text{eff}}^2 I_\omega^2 \sin^2 \left( \frac{\Delta k z}{2} \right)}{n_{2\omega}^2 n_\omega^2 c^3 \varepsilon_0 \Delta k^2}$$  \hspace{1cm} (3-40)

As we can see, as the light propagates in the crystal, the SHG power varies periodically as a $\sin^2$ function. The maximum value appears at:

$$\Lambda = \frac{\pi}{|\Delta k|} = \frac{\lambda_\omega}{4 |n_{2\omega} - n_\omega|}$$  \hspace{1cm} (3-41)

If the crystal polarization is flipped after every $\Lambda$, we have a spatially modulated $d_{\text{eff}}$, in this case the SHG power will keep increasing instead of dropping after the peak. In this situation, we can denote:

$$d_{\text{eff}} = \begin{cases} +d_{\text{eff}}, & z \in (2n + 1)\Lambda \\ -d_{\text{eff}}, & z \in 2n\Lambda \end{cases}$$  \hspace{1cm} (3-42)

Over a length $z=L=NA$, where $N$ is an integer, the SHG light field becomes:

$$E_{2\omega}(NA) = \frac{i\omega}{n_{2\omega} c_0} d_{\text{eff}} E_\omega \int_0^{NA} \exp(i\Delta k z) dz$$

$$= \frac{i\omega}{n_{2\omega} c_0 \Delta k} d_{\text{eff}} E_\omega \left[ \left( \int_0^n - \int_{\Lambda}^{2n \Lambda} + \int_{2n \Lambda}^{3n \Lambda} \cdots + (-1)^{N/2} \int_{(N-1)\Lambda}^{NA} \right) \exp(i\Delta k z) dz \right]$$

$$= \frac{\omega}{n_{2\omega} c_0 \Delta k} d_{\text{eff}} E_\omega \left[ (e^{i\Delta \omega A} - 1) - (e^{2i\Delta \omega A} - e^{i\Delta \omega A}) + (e^{3i\Delta \omega A} - e^{2i\Delta \omega A}) - \cdots + (-1)^{N-1} (e^{Ni\Delta \omega A} - e^{(N-1)i\Delta \omega A}) \right]$$

$$= \frac{\omega(e^{i\Delta \omega A} - 1)}{n_{2\omega} c_0 \Delta k} d_{\text{eff}} E_\omega \left[ 1 - e^{i\Delta \omega A} + e^{2i\Delta \omega A} - \cdots + (-1)^{N-1} e^{(N-1)i\Delta \omega A} \right]$$
\[
\frac{\omega (e^{i\Delta k} - 1)}{n_\omega c_0 \Delta k} d_{\text{eff}} E_\omega \frac{1 - (-1)^N e^{i\Delta k}}{e^{i\Delta k} + 1} \quad (3-43)
\]

So the light intensity can be calculated as:

\[
I_{2\omega} = \frac{1}{2} n_{2\omega} c_0 \varepsilon_0 E_{2\omega} E_\omega^* = \frac{2 \omega^2 \Lambda^2}{n_{2\omega}^2 n_\omega^2 c_0^2 \varepsilon_0^2} d_{\text{eff}}^2 I_\omega^2 \operatorname{sinc}^2 \left( \frac{\Delta k \Lambda}{2} \right) \left[ \frac{1 - (-1)^N \cos(\Delta k \Lambda)}{1 + \cos(\Delta k \Lambda)} \right]
\]

(3-44)

Under quasi phase matching condition $\Delta k \Lambda \rightarrow \pi$, the equation has a limitation of:

\[
I_{\text{QPM}} = \lim_{\Delta k \Lambda \rightarrow \pi} I_{2\omega} = \frac{2 \omega^2 \Lambda^2}{n_{2\omega}^2 n_\omega^2 c_0^2 \varepsilon_0^2} d_{\text{eff}}^2 I_\omega^2 \operatorname{sinc}^2 \left( \frac{\pi}{2} \right) N^2 = \frac{4}{\pi^2} \frac{2 \omega^2 L^2 d_{\text{eff}}^2}{n_{2\omega}^2 n_\omega^2 c_0^2 \varepsilon_0^2} I_\omega^2
\]

(3-45)

Comparing $I_{\text{QPM}}$ with quasi phase matching and $I_{\text{MAX}}$ with total phase matching, we have the relation:

\[
I_{\text{QPM}} = \frac{4}{\pi^2} I_{\text{MAX}}
\]

(3-46)

Fig. 3.3. SHG intensity with different phase matching conditions. QPM period $\Lambda$ is defined as in Eq. (3-41).
Chapter 4. LiNbO$_3$ Ridge Waveguide Modeling and Simulation

4-1. Refractive Index Sellmeier Equation

The refractive index Sellmeier equation of LiNbO$_3$ is as follows:

$$n_e^2 = 5.756 + 2.86 \times 10^{-6} f + \frac{0.0983 + 4.7 \times 10^{-8} f}{\lambda^2 - (0.2020 + 6.113 \times 10^{-8} f)^2} + \frac{189.32 + 1.516 \times 10^{-4} f}{\lambda^2 - 12.52^2} - 0.0132 \lambda^2$$

(4-1)

where parameter $f = (T - 24.5)(T + 570.82)$, $T$ is in the unit of Kelvin, and $\lambda$ is in the unit of microns.

Fig. 4.1. Refractive index dispersion curve of LiNbO$_3$ under room temperature (25°C).

4-2. Modeling of APE Ridge Waveguide

4-2-1. Refractive Index Profile after Proton-Exchanging

The ion exchanging process obeys 1-D diffusion law, and theoretically, its behavior can be well described using the diffusion equation. Considering one directional diffusing process, we have the well-known diffusing equation as follows:
\[
\frac{\partial \phi(y, t)}{\partial t} = \nabla \cdot [D(\phi, y) \nabla \phi(y, t)] = \frac{\partial}{\partial x} \left[ D(\phi, y) \frac{\partial \phi(y, t)}{\partial y} \right] \tag{4-2}
\]

where \( \phi \) is the density function of the diffusant, which varies with diffusing time \( t \) and diffusion length \( y \). \( D \) is the collective diffusion coefficient, which is also a function depending on the density of diffusant and the diffusion length.

However, the proton-exchange process is actually not a pure physical diffusion process. It contains chemical reaction as:

\[
\text{LiNbO}_3 + \text{PhCOOH} \leftrightarrow \text{Li}_x \text{H}_{1-x} \text{NbO}_3 + \text{PhCOOLi} \tag{4-3}
\]

This has added the complexity of the diffusion coefficient \( D \). As the reaction goes on, the original structure of \( \text{LiNbO}_3 \) start to change. X-ray and TEM scanning found that the proton-exchanged area contains various different phases. Mobility of diffusion ions is considerably affected by such changes\[35\]. Experimental studies have revealed that the diffusion coefficient is closely linked to local ion concentration. For example, the diffusion coefficient of hydrogen ion is found to be \( 10^3 \) times smaller than that of Li ion before 80\% of the Li ions are removed. However, as the diffusion goes on, when it passes this value, the diffusion coefficient of \( H^+ \) begins to rise drastically. In the contrary, there is no such change on the diffusion efficiency of Li ions.

Theoretical analysis shows that the collective diffusion coefficient in proton-exchange has the following expression\[35\]:

\[
D = \frac{D_H}{1 - (1 - \frac{D_H}{D_L}) \frac{C_H}{C_H + C_L}} \tag{4-4}
\]

\( D_H \) and \( D_L \) represent the diffusion coefficient of \( H^+ \) and \( Li^+ \); \( C_H \) and \( C_L \) represents the concentration of \( H^+ \) and \( Li^+ \) respectively. Here we will not go into the details of deducing this equation and calculating each value, since it is not relevant with our
study and eventually cannot deliver an exact value either. Instead, we use a numerical curve fitting approach to find a proper solution.

Experimental investigations have found that, after proton-exchange, a very thin layer with a step-like extraordinary index increase is formed on the crystal surface. And despite the complicated relations among those diffusion parameters, the PE layer thickness obeys the standard square root law\cite{40}. The index increase has also been carefully investigated\cite{41}.

\begin{equation}
  d = 2\sqrt{D_0 \exp(-Q/k_B T)t}
\end{equation}

\begin{equation}
  \Delta n_e(\lambda) = \sqrt{a_1 + \frac{a_2}{\lambda^2 - a_3^2}}
\end{equation}

In our study, proton-exchange is carried out using benzoic acid as proton source on the +z surface of LiNbO$\text{3}$ crystal at around 200°C. Under such condition the values for $D_0$ and $Q$ are $1.84 \times 10^9 \ \mu m^2/h$ and $0.974eV$\cite{40}, and constants $a_1, a_2, a_3$ are $7.43 \times 10^{-3}, 2.64 \times 10^{-3}$, and $0.336$ respectively\cite{41}.

![Graph](image)

Fig. 4.2. Proton-exchange depth vs. proton-exchange time on +z-LiNbO$\text{3}$ under 200°C.
Since a thin step-like hydrogen-rich layer is formed on the crystal surface after proton-exchange, the following annealing process can be treated as a semi-infinite diffusion problem. The solution can be described as:

\[ C(y,t) = \frac{C_0}{2} \left( \text{erf} \left( \frac{d + y}{2\sqrt{D_y t}} \right) + \text{erf} \left( \frac{d - y}{2\sqrt{D_y t}} \right) \right) \]  

(4-7)

In this equation, parameter \( C_0 \) is the initial hydrogen concentration in PE layer before annealing; \( d \) is the calculated PE layer thickness; and \( D_y \) is the collective hydrogen diffusion coefficient during annealing, which is calculated to be 1.56 \( \mu \text{m}^2/\text{h} \) in our case, according to earlier researches.

However, several papers have point out that this model does not agree with experimental results perfectly, the actual hydrogen distribution curve has a much longer tail penetrating into the substrate. As a result, several modified functions are used to improve the agreement, such as using two Gaussian functions or cosh function. Albeit, in our study, we find these models lack of theoretical support, and above all, such modification have almost no influence on the simulation result. So,
we will stick to the original model.

The refractive index profile of the waveguide is closely linked to the hydrogen concentration distribution, which can be described using the following equation\[35\]:

\[
n_{\text{APE}}(y, \lambda, t) = n_e(\lambda) + \Delta n_e(\lambda) \{1 - \exp[-\sigma \cdot C'(y, t)]\}
\]

\(n_e(\lambda)\) is the refractive index for extraordinary light in bulk LiNbO\(_3\), and \(\Delta n_e(\lambda)\) is the index change in PE layer; they can all be calculated using equations introduced earlier. \(C'(y, t)\) is the normalized hydrogen concentration profile, which is obtained by simply setting \(C_0=100\%\) (that makes \(C(0,0)=1\)) in function \(C(y, t)\). Parameter \(\sigma\) represents the index saturation rate, which is used to describe the nonlinear level of the relationship between hydrogen concentration profile and actual refractive index profile. Using the data obtained from our earlier researches\[46\], \(\sigma\) is calculated through an iterative process in order to find the best fitted solution. Its value is found to be 2.0 under the annealing condition of 1 atm \(O_2\), \(+z\) surface and 350°C.

![Fig. 4.4. Refractive index profile for annealed proton-exchanged \(+z\)-LiNbO\(_3\) at 1550 nm wavelength. Annealing time is fixed at 5 hours and 45 min, while PE time varies as indicated in figure.](image-url)
Fig. 4.5. Refractive index profile for APE +z-LiNbO$_3$ at 1064 nm wavelength. PE time varies as shown in figure, while annealing time is fixed at 5 hours and 45 min.

4-3. Waveguide Simulation

4-3-1. APE LiNbO$_3$ Ridge Waveguide Refractive Index Model

Using the results shown above, the refractive index data of proton-exchanged ridge waveguide is calculated and entered into commercial photonics simulation software, Apollo Photonic Solutions Suite (APSS), through which the numerical model is built and analyzed.

Since space is limited, we will only show two groups of simulated results, instead of 30 groups with different conditions. The selected conditions are:

1. for 1550 nm light: 120 min PE time, 5 hrs 45 min annealing time, 8 $\mu$m ridge width.
2. for 1064 nm light: 95 min PE time, 5 hrs 45 min annealing time, 6 $\mu$m ridge width.

Under these conditions, the fabricated waveguides have demonstrated the best performances, in terms of waveguide propagation loss and transmitted mode profile. The experiment details will be discussed in later chapters.
Fig. 4.6. (a) Index model of APE ridge waveguide designed for 1550 nm wavelength. (b) Corresponding vertical index distribution.

Fig. 4.7. (a) Index model of APE ridge waveguide designed for 1064 nm wavelength. (b) Corresponding vertical index distribution.
4-3-2. Simulation Results

The simulation is run on APSS, the optical mode is calculated, and the horizontal and vertical profiles across the beam center peak are measured. The results are shown in the following figures.

![Optical Field](image1)

(a) Color view, and (b) contour view.

Fig. 4.8. Optical field of LiNbO₃ ridge waveguide designed for 1550 nm wavelength.

(a) Color view, and (b) contour view.

![Normalized intensity vs. X-axis](image2)

(a)
Fig. 4.9. (a) Horizontal and (b) vertical light field profile across Fig. 4.8 beam center. 
Here the beam center is taken as the zero point for both two graphs.

Fig. 4.10. Optical field of LiNbO₃ ridge waveguide designed for 1064 nm wavelength. 
(a) Color view, and (b) contour view.
Fig. 4.11. (a) Horizontal and (b) vertical light field profile across Fig. 4.10 beam center. In both two graphs, the beam center is taken as the zero point.
Chapter 5. Fabrication of Proton-Exchanged Ridge Waveguides

5-1. General Description of Fabrication Process

The fabrication process contains three major steps, namely proton-exchange, annealing and diamond blade dicing. All waveguides are formed on the +z surface of the crystal. Two groups of waveguide working at 1550 nm and 1064 nm are developed separately, but the fabrication processes are almost the same.

In order to obtain low loss waveguide with a single transmission mode, the proton-exchange condition and the ridge width must be optimized. In our earlier studies, we have found that the annealing time is not as sensitive as the proton-exchange time\cite{46}, so an empirical value of 5 hrs 45 min is used in the experiment. To find the best fabrication condition, 5 different proton-exchange time and 3 different ridge widths have been investigated for each wavelength. In order to
improve data accuracy, multiple samples with the same parameters are made and tested

Taking waveguides designed for 1550 nm light for example, the fabrication procedure is, first, a 3-inch LiNbO₃ wafer is separated into 5 identical pieces, then 5 different proton-exchange conditions with the same annealing time are applied to each piece. After that, each piece is separated into 3 parts, and each part is further divided into 2 sections with 1 cm length difference. At last, 4 ridge waveguides are diced on each section. The ridge widths are the same for all sections on the same part, but 3 parts have 3 different ridge widths. (See Fig. 5.2.)

As a result, we have 120 waveguide samples for tests at 1550 nm wavelength. 15 different fabrication conditions are included; each 8 waveguides have the same fabrication condition; 4 of them are 1 cm longer than the other 4 waveguides. Similarly, for 1064 nm wavelength, we will need another 120 waveguides. After testing these 240 waveguide samples, the best fabrication conditions can then be decided. The whole fabrication flow chart is shown in Fig. 5.2.

5-2. Proton-Exchanging and Annealing

5-2-1. Benzoic Acid Proton-Exchanging

Each LiNbO₃ sample is placed in a ceramic crucible, and then 50 g 99.9% benzoic acid is added into the crucible. After that, the crucible is transferred into a furnace with automatic ventilation and programmable temperature control. Proton-exchange temperature is kept at 185°C after a 65 min ramp time. After a certain time, the furnace heater is turned off, and the crucible is naturally cooled down over night.
Fig. 5.2. General workflow for APE LNO ridge waveguide optimization.

| Ridge 1 Long 1 (APER0116L01) | Dicing | Separation |
| Ridge 1 Long 2 (APER0116L02) |        |            |
| Ridge 1 Long 3 (APER0116L03) |        | Ridge 1 Long (APER0116L) |
| Ridge 1 Long 4 (APER0116L04) |        |            |
| Ridge 1 Short 1 (APER0116S01) |        | Ridge 1 Short (APER0116S) |
| Ridge 1 Short 2 (APER0116S02) |        |            |
| Ridge 1 Short 3 (APER0116S03) |        |            |
| Ridge 1 Short 4 (APER0116S04) |        |            |
| Ridge 2 Long 1 (APER0112L01) |        |            |
| Ridge 2 Long 2 (APER0112L02) |        | Ridge 2 Long (APER0112L) |
| Ridge 2 Long 3 (APER0112L03) |        |            |
| Ridge 2 Long 4 (APER0112L04) |        |            |
| Ridge 2 Short 1 (APER0112S01) |        | Ridge 2 Short (APER0112S) |
| Ridge 2 Short 2 (APER0112S02) |        |            |
| Ridge 2 Short 3 (APER0112S03) |        |            |
| Ridge 2 Short 4 (APER0112S04) |        |            |
| Ridge 3 Long 1 (APER0108L01) |        |            |
| Ridge 3 Long 2 (APER0108L02) |        | Ridge 3 Long (APER0108L) |
| Ridge 3 Long 3 (APER0108L03) |        |            |
| Ridge 3 Long 4 (APER0108L04) |        |            |
| Ridge 3 Short 1 (APER0108S01) |        | Ridge 3 Short (APER0108S) |
| Ridge 3 Short 2 (APER0108S02) |        |            |
| Ridge 3 Short 3 (APER0108S03) |        |            |
| Ridge 3 Short 4 (APER0108S04) |        |            |

Separation

Annealing

Proton-Exchange

Similar procedures

3 inch LN
5-2-1. Thermal Annealing

After proton-exchange, the melt benzoic acid is removed using hot acetone. The sample is taken out and washed several times using ethanol and DI water. Then the sample is transferred into an open titanium crucible, and placed into a three-section furnace for annealing. The temperature is precisely controlled at 355°C, and the annealing time is fixed at 5 hrs 45 min. Flowing wet O$_2$ is used to suppress Li$_2$O out-diffusion during the annealing. All detailed parameters used at 1550 nm and 1064 nm wavelength are listed in the following tables.

![Temperature Curve](image-url)

Fig. 5.3. Temperature curve of (a) proton-exchange and (b) annealing process.
Table. 5.1. Proton-exchange conditions for LiNbO$_3$ ridge waveguides designed for 1550 nm wavelength.

<table>
<thead>
<tr>
<th>Sample group</th>
<th>Time</th>
<th>APE01</th>
<th>APE02</th>
<th>APE03</th>
<th>APE04</th>
<th>APE05</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ramp</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1hr5min</td>
</tr>
<tr>
<td>Soak</td>
<td></td>
<td>1hr 45min</td>
<td>1hr 53min</td>
<td>2 hrs</td>
<td>2hr 7min</td>
<td>2hr 15min</td>
</tr>
<tr>
<td>Cool down</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>natural cooling</td>
</tr>
</tbody>
</table>

Table. 5.2. Proton-exchange conditions for LiNbO$_3$ ridge waveguides designed for 1064 nm wavelength.

<table>
<thead>
<tr>
<th>Sample group</th>
<th>Time</th>
<th>APE06</th>
<th>APE07</th>
<th>APE08</th>
<th>APE09</th>
<th>APE10</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ramp</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1hr5min</td>
</tr>
<tr>
<td>Soak</td>
<td></td>
<td>1hr 20min</td>
<td>1hr 28min</td>
<td>1hr 35min</td>
<td>1hr 42min</td>
<td>1hr 50min</td>
</tr>
<tr>
<td>Cool down</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>natural cooling</td>
</tr>
</tbody>
</table>

5-3. Precise Diamond Blade Dicing

5-3-1. Selecting the Right Blade

To make a ridge waveguide by dicing, selecting the right blade is the most important issue. There are countless commercial products in the market, but a suitable one for dicing LiNbO$_3$ with excellent smoothness is still hard to find. First of all, most commercial products are not designed for such a delicate operation, and second, LiNbO$_3$ is a hard and brittle material with no clear cleavage plane, which can easily
generate chipping and cracking. Diamond blade dicing is a complicated chemical and mechanical science, which also involves lots of experimental and empirical knowledge. We cannot fully explore it in this study, so only several crucial parameters are discussed in the following paragraphs.

1. Mesh size — As we have mentioned, the diamond dicing blade is made of diamond particle embedded in a bonding matrix. Mesh size is used to describe the size of diamond particles bonded in the blade. Generally, smaller diamond particle size generates better cutting quality, less chipping and smooth kerfs. However, at the same time, the cutting speed drops, the blade becomes hard to maintain. Since the dicing quality is directly linked to waveguide propagation loss, such trade-off must be made. A mesh size higher than #4000 is necessary to avoid excessive chipping for optical materials.

<table>
<thead>
<tr>
<th>U.S. Standard Mesh</th>
<th>Inches</th>
<th>Microns</th>
</tr>
</thead>
<tbody>
<tr>
<td>#100</td>
<td>0.0059</td>
<td>150</td>
</tr>
<tr>
<td>#200</td>
<td>0.0030</td>
<td>74</td>
</tr>
<tr>
<td>#325</td>
<td>0.0017</td>
<td>43</td>
</tr>
<tr>
<td>#400</td>
<td>0.0015</td>
<td>37</td>
</tr>
<tr>
<td>#1200</td>
<td>0.0005</td>
<td>12</td>
</tr>
<tr>
<td>#2400</td>
<td>0.0002</td>
<td>6</td>
</tr>
<tr>
<td>#4800</td>
<td>0.0001</td>
<td>2</td>
</tr>
</tbody>
</table>

Table. 5.3. Standard mesh-inch-micron conversion table.

2. Bond Hardness — There are in general three kinds of bonding matrixes: Sintered bond, electroplated bond, and resin bond. From first one to the last, the bonding hardness is changing from high to low. Soft bonding matrixes will make the
diamond particles more flexible during dicing, but also cause them easy to wear off; harder bonding increases the cutting strength and blade life, but more easily to generate a scratched finish. LiNbO$_3$ is a brittle crystalline material with moderate hardness, and theoretically, in order to obtain best cutting quality, a resin bond blade will be the best choice. However, usually a resin bond blade cannot be made very thin, because in that case the blade cannot survive under the dicing strength. Therefore, the electroplated bond is also a good choice for both thin kerf and good dicing quality.

3. Blade thickness – As mentioned above, a thin dicing kerf is expected for good cutting finish, because in this case, less dicing strength is required, less heat is generated, and better quality can be obtained. The trade off is shorter blade life, and of course, the manufacture cost is higher, and handling becomes difficult.

5-3-2. Formation of Ridge Waveguide

A good diamond blade does not ensure high quality ridge waveguide. Proper dicing parameters must be set. In order to produce chipping-free ridge and smooth sidewalls, dicing strength and local vibration must be minimized. First, the blade must be installed precisely; the blade plane must be precisely perpendicular to the spindle. Debris on the spindle flange greatly affects the cutting quality. The spindle speed, the material feeding speed, and cooling water pressure must be set to a proper level. The different finishing quality using different parameters are obvious under the microscope.

The sample also has to be mounted on the dicing saw exactly, in order to avoid excessive cutting depth variation along the dicing direction. After adjustment, the depth variation across a 2 cm long LiNbO$_3$ chip is controlled under 1 $\mu$m in our experiment.
Fig. 5.4. Optical image of LiNbO$_3$ ridge waveguides diced with (a) non-optimized parameter, and (b) optimized parameter. The ridges are around 16 μm wide.

5-3-3. Waveguide Facet Polishing

The waveguide facet cannot be processed before ridge dicing, since the ridge ends are usually rather rough and need to be cut away. At first, a traditional method is used to polish the facet. The chip ends are first polished using 0.5 μm diamond slurry, and then using nano SiO$_2$ polishing solution. However, we found the polished facet even get worse, the facet is totally damaged. (See Fig. 5.5.)

As a result, a dicing method is used to process the waveguide facets. This method is similar to ridge dicing, except the cutting depth is much larger. This allows easy coupling between the waveguide and optical fiber. The remaining roughness on the facet is similar to ridge sidewall.
Fig. 5.5. LiNbO$_3$ ridge waveguide facet polishing results. Photo (a) shows the damaged waveguide facet after traditional optical polishing. Photo (b) shows the facet using diamond blade dicing, showing no mechanical damage, although the waveguide size is much smaller than (a).
Chapter 6. Characterization Annealed Proton-Exchanged LiNbO$_3$ Ridge Waveguide

6-1. Geometric Appearance

The fabricated ridge waveguides are first examined under optical microscope and SEM. The following pictures show a chip containing three ridge waveguides with 10 μm width. The smooth sidewalls ensure least light scattering loss, and the well-polished facet enables easy coupling from optical fiber. The tilted ridge shape is caused by the diamond blade, which has an asymmetric profile at the tip.

Dicing quality can be further improved if better dicing blades with smaller diamond particle size are used. The alignment accuracy of current dicing saw is quite limited, 2 μm at X and Y direction, and 1 μm at Z direction. The shape of the ridge waveguide can be better controlled if finer machinery is available.

Fig. 6.1. Microscopic picture of a ridge waveguide chip. (a) End part. (b) Middle part.
Fig. 6.2. SEM photo of a ridge waveguide chip. (a) (b): left view; (c) (d) right view.

6-2. Waveguide Propagation Loss

6-2-1. Instrument Setup

All measurements are made with the following setup:

Fig. 6.3. Instrument setup of waveguide measurement.
Light is coupled out from a fiber laser source using a SM fiber, and directly coupled into the ridge waveguide. Then the waveguide output is focused through a 25× objective lens, an attenuator, a split mirror, then into the power meter. Infrared camera is used to record transmitted mode profile, which will be discussed later. The waveguide propagation loss is calculated by comparing the total insertion loss of two waveguides with the same fabrication parameters but different lengths.

\[
\text{Propagation Loss} = \frac{\text{Short WG Output} - \text{Long WG Output}}{\text{Long WG Length} - \text{Short WG Length}} \quad (6-1)
\]

6-2-2. Waveguide Propagation Loss at 1550 nm Wavelength

The best value is 0.17 dB/cm, which belongs to a group that has 8 μm ridge width and 2 hrs PE time. The second best value is 0.32 dB/cm, which comes with 16 μm ridge width and 2 hrs 7 min PE time. The full values are listed in the table below. From these data and the plot, we can conclude that a proton-exchange time of around 2 hrs 7 min yield the best results.

<table>
<thead>
<tr>
<th>PE Time</th>
<th>1hr45min</th>
<th>1hr52min</th>
<th>2 hrs</th>
<th>2 hrs7min</th>
<th>2 hrs15min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ridge Width</td>
<td>16 μm</td>
<td>16 μm</td>
<td>16 μm</td>
<td>16 μm</td>
<td>16 μm</td>
</tr>
<tr>
<td>Propagation Loss(dB/cm)</td>
<td>3.88</td>
<td>3.41</td>
<td>1.08</td>
<td>0.32</td>
<td>1.20</td>
</tr>
</tbody>
</table>

(6-1-1)

<table>
<thead>
<tr>
<th>PE Time</th>
<th>1hr45min</th>
<th>1hr52min</th>
<th>2 hrs</th>
<th>2 hrs7min</th>
<th>2 hrs15min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ridge Width</td>
<td>12 μm</td>
<td>12 μm</td>
<td>12 μm</td>
<td>12 μm</td>
<td>12 μm</td>
</tr>
<tr>
<td>Propagation Loss(dB/cm)</td>
<td>2.16</td>
<td>1.78</td>
<td>0.62</td>
<td>0.62</td>
<td>0.60</td>
</tr>
</tbody>
</table>

(6-1-2)
### Table 6.1. Measured waveguide propagation loss at 1550 nm wavelength.

<table>
<thead>
<tr>
<th>PE Time</th>
<th>1hr45min</th>
<th>1hr52min</th>
<th>2 hrs</th>
<th>2 hrs7min</th>
<th>2 hrs15min</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ridge Width</td>
<td>8 μm</td>
<td>8 μm</td>
<td>8 μm</td>
<td>8 μm</td>
<td>8 μm</td>
</tr>
<tr>
<td>Propagation Loss (dB/cm)</td>
<td>2.32</td>
<td>1.46</td>
<td>0.17</td>
<td>0.67</td>
<td>1.19</td>
</tr>
</tbody>
</table>

(6-1-3)

![Graph showing propagation loss vs. proton-exchange time at 1550 nm wavelength.](image)

Fig. 6.4. Propagation loss vs. proton-exchange time at 1550 nm wavelength.

#### 6-2-3. Waveguide Propagation Loss at 1064 nm Wavelength

The lowest propagation loss for 1064 nm light is 0.30 dB/cm, which have a PE time of 1 hr 35 min and ridge width of 6 μm. The waveguide fabricated with 1 hr 50 min PE time also have a 0.30 dB/cm loss, however, we will demonstrate later that this condition is not desirable. Comparing with waveguides fabricated for 1550 nm light, as the wavelength becomes shorter and fabrication requirement becomes higher, the lowest waveguide propagation loss obtainable increases obviously.
Table 6.2. Measured waveguide propagation loss at 1064 nm wavelength.
Fig. 6.5. Propagation loss vs. proton-exchange time at 1064 nm wavelength.

6-3. Transmitted Mode Profiles

The transmitted mode profile of each waveguide is recorded by CCD camera. As the PE time and ridge width increase, the fabricated ridge waveguides begin to show multimode propagation profiles when light is launched into the waveguides.

Since there is no enough space to show every picture here, we will only demonstrate the measured results from the waveguides, which have lowest propagation loss. For the selected condition at 1550 nm wavelength, the acquired data still suggest that the waveguides are working at single mode transmission conditions.

6-3-1. Near-Field Mode Profile at 1550 nm Wavelength

The measured result shown below has a corresponding fabrication condition of 120 min PE time, 5 hrs 45 min annealing time and 8 μm ridge width. Both the infrared photo and measured intensity profiles suggest that the ridge waveguide has tight light confinement at three directions.
Fig. 6.6. Mode profile of waveguide designed for 1550 nm wavelength. (a) CCD image, (b) horizontal profile and (c) vertical profile.

6-3-2. Near-Field Mode Profile at 1064 nm Wavelength

The mode profiles of waveguide fabricated with 1 hr 35 min PE time and 1 hr 50 min PE time are shown in the following figures. The later one shows multiple transmission modes, which is not desirable in our case.
Fig. 6.7. Mode profile of waveguide designed for 1064 nm wavelength. (a) CCD image, (b) horizontal profile and (c) vertical profile.

6-4. Simulation & Experiment Comparison

Comparing the measured near-field mode profiles of waveguides fabricated for both 1550 nm and 1064 nm wavelength, and the simulated results obtained earlier, we have found excellent agreements between each other. The vertical profiles almost
overlap with each other, while the longitudinal ones deviate a little bit from one another. We presume that this deviation originates from instrumental limitation during dicing. The minimum alignment accuracy on the step motor of the dicing saw is 2 μm; after careful hand alignment, this value could be reduced to 1 μm, however, it is inevitable that the actual dicing line still have some deviation from the panel reading.

Fig. 6.8. Comparison between measured and calculated mode profile at 1550 nm.
Fig. 6.9. Comparison between measured and calculated mode profile at 1064 nm.
Chapter 7. Quasi-Phase Matched Second Harmonic Generation Using Annealed Proton-Exchanged LiNbO₃ Ridge Waveguide

7-1. Experiment Description

Using APE PPLN ridge waveguides, considerable conversion efficiency can be achieved through QPM SHG. Using 1064 nm light and fundamental source, 532 nm green light can be generated conveniently. Since miniaturized green light source is highly desirable in many applications such as medical instrumentation and advanced display.

In this chapter, our final objective is to generate green light with a power of more than 100 mw, using a single APE PPLN ridge waveguide. The maximum achievable output, SHG conversion efficiency and output beam quality will be assessed.

7-2. Fabrication Procedures

The general fabrication procedures are similar to what introduced in earlier chapters. In order to enhance the device performance under high power density, MgO doped LiNbO₃ is used instead of congruent ones. As a result, the proton-exchange parameter is a little bit different to what described earlier. According to our empirical results, MgO doped LiNbO₃ need an additional 30min to reach the same propagation loss level as congruent LiNbO₃, and the annealing time is 30min longer.

The PPLN chip is also optimized to carry out SHG at near room temperature. Since proton-exchange will result in a remarkable refractive index increase, the polling period must be reduced to compensate such increase. The following table sums up the experiment parameters. Waveguides with ridge width of 4, 6, 8, 10 and 12 µm are diced and tested.
7-3. Green Light Generation

7-3-1. Instrument Setup

Light is coupled out from a 1063.8 nm high power fiber laser through PM fiber. The polarization direction is adjusted to fit the TM polarized APE ridge waveguide. The APE MgO:PPLN ridge waveguide samples are placed on a copper holder. Infrared radiation is converted into green light after passing through the ridge waveguide. Temperature controller with 0.1 °C accuracy is applied on the waveguide holder. The output light is focused by a 25× objective lens and into a power meter. The green light power and total output power is measured with/without the green filter.

![Instrument setup for SHG experiment.](image)

The measured SHG output and conversion efficiency curve are shown in the following figure. Light power loss caused by the objective lens and filter has already been compensated.
Fig. 7.2. (a) 1063.8 nm-531.9 nm SHG output power, and (b) conversion efficiency.

The 6 μm wide ridge waveguide has demonstrated more than 125 mw green output under 250 mw total output power; 8 μm ridge waveguide is also able to produce more than 100 mw green light. The highest conversion efficiency 53.2% is
achieved by the 6 μm ridge waveguide under 216 mw total output. Conversion efficiency of waveguides with 4, 6, 8 μm width is significantly higher than 10 and 12 μm wide ridge waveguides. The normalized conversion efficiency for 6 μm wide ridge waveguide is 127%/W·cm² at 100 mw total output power, and becomes higher under low power operation. To the best of our knowledge, this is the highest value ever achieved using APE waveguide for green light generation,

7-3-3. Output Mode Profile

The waveguide output beam is focused and measured by a CCD camera, using the similar setup as introduced in earlier chapters. Near-field mode profiles for both 1063.8 nm and 531.9 nm light are recorded.
Fig. 7.3. Mode profile of (a) 1063.8 nm and (b) 531.9 nm light output from 6 μm wide APE MgO:PPLN ridge waveguide. (a-1) Infrared photograph, (a-2) horizontal profile and (a-3) vertical profile.

7-3-4. Existing Problems

Although theoretically the 4 μm wide ridge waveguide should have the highest output and conversion efficiency, in our experiment, it did not show better performance than 6 μm or 8 μm wide ridge waveguides. Its conversion ability begins to cease under more than 150 mw pumping power, and serious output decrease occurred with more than 200 mw pumping. Similarly, for 6 μm and 8 μm wide ridge waveguides, as fundamental input keeps growing, the SHG conversion efficiency growing rate becomes slower, and finally begins to drop.

The problem is two-fold, first as the waveguide width decreases, fabrication becomes more rigid, and a narrower ridge tends to suffer higher propagation loss, especially for shorter SHG wavelength. Such a loss increase will counter-act with power density increase and result in no improvement. However, since SHG conversion efficiency grows as the fundamental power increases, while waveguide loss stays the same, the 4 μm wide ridge waveguide still show higher conversion efficiency than 6 μm and 8 μm wide ridge waveguides under 110-170 mw total output.
Secondly and more importantly, as the pumping power increases, heat generation becomes severe inside the waveguide. Under 200 mw total light power, the power density inside a 6 μm wide ridge waveguide exceeds 0.6 MW/cm². Thermal conductivity of LiNbO₃ is very low, no more than 4 W/mK. Above all, the ridge waveguide height is only several microns, but it is built on a 500 μm thick crystal. Heat dissipation becomes very difficult under such high power density and poor thermal conductivity. From the bottom side, the temperature controller cannot precisely adjust the temperature inside the waveguide through a medium 100 times thicker. From the top side, although natural convection can take away a large quantity of heat, but it easily causes temperature non-uniformity over the 1.6 cm long chip.

In conclusion, under high power density operation, the waveguide temperature must be controlled more precisely. Improving temperature uniformity across the whole chip and cutting down thermal resistance between waveguide and controller are proper measures to achieve better performance.

7-4. Enhancing Device Performance Using Advanced Packaging

7-4-1. Description of Device Structure

In order to improve device performance under high power input, a sandwiched micro channel packaging is designed and fabricated. The schematics are shown in the following figure.

At the front side, the whole waveguide is covered under a thermal conductive lid, which has a carved out micro channel. The micro channel will protect the fragile waveguide from scratching or dust contamination, and most importantly forms a comparatively stable environment free of natural convection disturbance. At the backside of the chip, its total thickness is significantly cut down into several tens of microns, in order to get higher thermal conductivity. In addition, to protect the
backside of the chip, a supporting substrate with high thermal conductivity is added. By using this packaging method, the thermal conductivity of the device can be greatly enhanced. For example, if the LiNbO$_3$ crystal thickness is thinned down to 100 μm, considering 2-D thermal dissipation model, the total thermal conductivity can be 25 times higher than an unpacked waveguide chip.

![Diagram of LiNbO$_3$ ridge waveguide packaging with enhanced thermal management](image)

Fig. 7.4. Cross-section schematics of LiNbO$_3$ ridge waveguide packaging with enhanced thermal management. The covering chip and supporting chip can be any material with high thermal conductivity and easy to process.

7-4-2. Device Fabrication

The experiment is carried out on a 1.2 cm long waveguide chip that contains three guides. First, two 500 μm thick Si wafer pieces are prepared with the same size as the chip. Then a 35 μm deep micro channel is diced out on one Si chip; the channel width is large enough to cover all three waveguides. After that, the waveguides are aligned to the micro channel and two chips are combined using a thin layer of strong adhesive epoxy. Next step, the chip is flipped over and mounted on the dicing saw for thinning down. The thickness of LiNbO$_3$ chip is reduced to about 90 μm. Finally, the thinned waveguide chip is glued to another Si supporting chip by thermal conductive paste.
Fig. 7.5. Fabrication process of packaged LiNbO$_3$ ridge waveguide with improved thermal management.
Fig. 7.6. Microscope pictures of packaged LiNbO$_3$ ridge waveguide, showing the chip facet and micro channel.

7-4-3. Experimental Results and Comparison

The waveguide chip is tested before and after packaging using the same method as described in the earlier chapter. By comparing the results, significant improvement is observed under high power pumping. 180 mw green output is achieved with 500 mw total power, where the top conversion efficiency is 36% (equals to 320 mw green output and 64% conversion efficiency if 1.6 cm chip is used).

However, there are still questions remaining. For example, the conversion efficiency stops growing after 36%, the curve shows a typical shape of efficiency saturation. In this case, the turning point is around a power density of 0.8 MW/cm$^2$. Currently there is no sufficient evidence to explain it, only presumably caused by green-induced infrared absorption. Further study is needed to reveal its nature.
Fig. 7.7. (a) SHG green output power and (b) conversion efficiency of packaged APE MgO:PPLN ridge waveguide. The waveguide is 8 µm wide and 1.2 cm long. Device performance is greatly enhanced at high power range.
Fig. 7.8. Temperature tuning curve of packaged waveguide under 100 mw total output.
Chapter 8. Conclusions

In this study, the existing LiNbO$_3$ waveguide fabrication techniques are reviewed in detail; several important and most popular processes such as Ti diffusion, annealed proton-exchange and precise machining are fully elaborated. Based on the existing technologies, a novel type of annealed proton-exchanged ridge waveguide with high height to width ratio is proposed. By combining such waveguide and periodically polling, quasi-phased matched second harmonic generation can be carried out with considerably high conversion efficiency.

Theories regarding QPM SHG, proton-exchange, and thermal diffusion are carefully reviewed. Refractive index model at both 1550 nm and 1064 nm wavelengths are successfully built by referring to earlier experimental results; and the waveguide propagation mode is calculated using commercial photonics simulation software.

The proposed APE LiNbO$_3$ ridge waveguide is successfully fabricated by combining annealed proton-exchange and precise diamond blade dicing. In order to find the best fabrication parameters both for 1550 nm and 1064 nm wavelength, 15 different conditions are chosen, and 120 waveguide samples are made and tested for each wavelength. Under the selected conditions, the APE ridge waveguides have demonstrated single transmission mode and a propagation loss as low as 0.17 dB/cm for 1550 nm and 0.3 dB/cm for 1064 nm wavelength. The experimental results also show good consistency with the simulated results.

APE ridge waveguide is also built on MgO:PPLN wafer successfully. QPM SHG at around 532 nm has been demonstrated with considerably high conversion efficiency. The 6 μm wide 1.6 cm long ridge waveguide has achieved more than 125 mw green light output under 250 mw total output power, and the top conversion efficiency 53.2%. However, under high pumping power the waveguide green output begins to
drop, due to thermal non-uniformity over the whole chip. In order to alleviate this situation, an advanced packaging design using micro channel sealing and backside thinning down methods is proposed and realized. Improvements are obvious; the waveguide’s high power performance is greatly improved. However, there are still several remaining questions under high power density, which need to be explored in the future.

Based on this well-established platform of LiNbO₃ ridge waveguide, various experiments can be conducted, and several useful optical devices could be built. For example, it can be used as nonlinear wavelength convertor for mid-infrared generation and compact blue laser, high quality modulators, and possibly optical sensors. The potentials have not yet been fully investigated.
References


[2]. T. R. Volk et al, Non-photorefractive LiNbO₃:Mg as the effective material for the nonlinear optics. Ferroelectrics 95, 121 (1989).


[6]. Xijun Li et al, Micro and Nano Domain Engineering in LiNbO₃ and LiTaO₃ by Focused Charge Beam, CLEO/QELS, paper JTuD50, 2006.


[26]. H. Hu et al., Large area, crystal-bonded LiNbO₃ thin films and ridge waveguides of high refractive index contrast,” Topical Meeting “Photorefractive Materials, Effects, and Devices – Control of Light and Matter”, 2009.


[39]. O. Gayer et al., “Temperature and wavelength dependent refractive index equations for MgO-doped congruent and stoichiometric LiNbO3,” Appl. Phys. B.
vol.91, 343 (2008).


