Waveguides in single-crystal lithium niobate thin film by proton exchange

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Abstract: The proton exchanged (PE) planar and channel waveguides in a 500 nm thick single-crystal lithium niobate thin film (lithium niobate on insulator, LNOI) were studied. The mature PE technique and strong confinement of light in the LN single-crystal thin film were used. The single mode and cut-off conditions of the channel waveguides were obtained by finite difference simulation. The results showed that the single mode channel waveguide would form if the width of the PE region was between 0.75 μm and 2.1 μm in the β4 phase. The channel waveguide in LNOI had a much smaller mode size than that in the bulk material due to the high-refractive-index contrast. The mode size reached as small as 0.6 μm2 in simulation. In the experiment, the refractive index and phase transition after PE in LNOI were analyzed using the prism coupling method and X-ray diffraction. Three different width waveguides (5 μm, 7 μm and 11 μm) were optically characterized. Near-field intensity distribution showed that their mode sizes were 3.3 μm2, 5 μm2 and 7 μm2. The propagation losses were evaluated to be about 16 dB/cm, 12 dB/cm and 11 dB/cm, respectively. The results indicate that PE is a promising method for building more complicated photonic integrated circuits in single-crystal LN thin film.

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1. Introduction

In recent years, high-refractive-index contrast, single-crystal lithium niobate (LN) thin film on a low refractive index SiO2 cladding layer (lithium niobate on insulator, LNOI) or other substrates, has been fabricated using crystal ion slicing and other wafer bonding technologies and is widely used [1–5]. The high-refractive-index contrast between LN and SiO2, resembling silicon on insulator (SOI), enables good confinement and strong guiding of light, thus a high-density photonic integrated circuit (PIC) is expected. Compared with SOI-based optical devices, LN has excellent electro-optic, nonlinear and acousto-optic properties, and can be
easily doped with rare-earth ions to prepare laser material [6]. Various photonic devices have been reported in LN thin films, such as photonic crystals [7], electro-optical modulators [8,9], micro-ring resonators [10,11], microdisk resonators [12,13] and even heterogeneous LN photonic devices [14]. Waveguide is the fundamental structure in building photonic devices in LN thin film, however, until now, most of the waveguides in LN thin film have been formed using the dry etching process [15]. Since LN is a hard material to etch, the dry etching method is relatively difficult. Furthermore, the rough dry-etched sidewall leads to scattering loss of light [16].

Compared with dry etching, proton exchange (PE) is a low-cost and mature technology that has been widely used for fabricating optical devices in lithium niobate bulk material [17]. The PE waveguide has a smooth boundary, in contrast to the rough etched sidewall. During PE, the extraordinary refractive index \( n_e \) increases and a waveguide structure forms. The phase transition, optical properties, channel waveguides in PE bulk LN have been intensively studied [18–20]. The photonic devices based on PE bulk LN, such as photonic crystal slab [21], temperature sensor [22], second harmonic generation in buried and ridge waveguides [23,24] and more complicated devices using PE waveguide have been reported [25]. However, due to the small increase in refractive index in the PE region, light is weakly confined, which blocks the development of ultra-compact PIC.

PE seems to be a suitable solution to build PIC on LNOI because the rough sidewall can be avoided and one can take advantage of the strong confinement of light in LN thin film. Therefore, it is necessary to study the PE process and the PE waveguides in LNOI. More complicated optical devices, such as electro-optical modulator and second harmonic generator, based on PE in LNOI, can then be explored.

In this paper, the PE planar and channel waveguides in LNOI were studied by simulation and experimentation. The single mode and cut-off conditions and the mode profile of the channel waveguide in LNOI were simulated using the commercial software “Lumerical: Mode Solution”, based on a full-vectorial finite difference method [26,27]. In the \( \beta_4 \) phase, the channel waveguide was single mode when the PE width was between 0.75 \( \mu \)m and 2.1 \( \mu \)m. A much smaller mode size of 0.6 \( \mu \)m\(^2\) were obtained compared with the conventional PE waveguide. The change in refractive index and the PE LNOI film thickness were measured via the prism coupling method. A crystalline phase change was found by the X-ray diffraction. The channel waveguides on LNOI (500 nm thick LN film) were formed by the photolithography process. Three channels with widths of 5 \( \mu \)m, 7 \( \mu \)m and 11 \( \mu \)m were characterized. The mode sizes of these channels were 3.3 \( \mu \)m\(^2\), 5 \( \mu \)m\(^2\) and 7 \( \mu \)m\(^2\) and the propagation losses were evaluated to be about 16 dB/cm, 12 dB/cm and 11 dB/cm, respectively by the Fabry-Perot resonator method. The realization of PE channel waveguide could lead to more advanced and complicated integrated optical devices and circuits based on LNOI.

2. Simulation

All of the LN films used in this study, in both simulations and experiments, were Z-cut. Since PE causes an \( n_e \) increase, only the quasi-TM mode was studied. The schematic cross-section of the PE channel waveguide in LNOI is shown in the right inset of Fig. 1. A single-crystal LN thin film was bonded to a SiO\(_2\) layer on a LN substrate. Then, a part of the LN thin film was proton exchanged and the waveguide was formed by the \( n_e \) increase. The index-increased region, marked by “PE,” was assumed to have a step-like profile [28]. This structure belonged to a type of waveguide called a “strip-loaded waveguide” [29], although there were some differences. In this study, the strip was the PE region of the film instead of on the surface of the film. In such a waveguide, the cut-off width of the strip was zero for the fundamental mode [28]. In the simulation of the PE channel waveguide in LNOI, a zero cut-off width (no matter how narrow the PE region was, there was always a guided mode) was indeed obtained. However, when the waveguide dimension was small enough and got smaller, more and more light energy of the mode distributed outside the waveguide core (PE region). Thus, in this
paper, the cut-off width was defined as a minimum width at which 50% of the light energy was confined to the PE region. A full-vectorial finite difference method was used in the simulation and the result is shown in Fig. 1. The lines and symbols in black are the single mode and fundamental mode cut-off conditions with a refractive increase of 0.09 at 1539 nm, which was a measured value in this work. The X-axis is the thickness of the LN thin film and the Y-axis is the width of PE region. For example, for a 0.65 μm thick LN layer, the cut-off width was about 0.75 μm; when the waveguide width increased to above 2.1 μm, a second mode would appear. As a result, a single mode PE waveguide should have a width between 0.75 and 2.1 μm. The intensity distributions of the fundamental mode “a” and the second mode “b” were calculated and are shown in the left inset of Fig. 1 for the geometries labeled as “a” and “b” on the curves of Fig. 1.

Fig. 1. Top: single mode and fundamental mode cut-off conditions for the PE waveguide with Δn = 0.09 simulated at the wavelength of 1550 nm. The cut-off thickness (0.2 μm) was the minimum LN film thickness above which the fundamental quasi-TM mode of the planar waveguide was supported. Inset: near-field intensity distributions of the fundamental mode and the first higher order mode in the PE channel waveguides with the corresponding dimension labeled as a, b on the curves.

To enable the development of ultra-compact PIC and to strengthen the nonlinear and electro-optical effect, a small mode size is expected. Figure 2 shows the simulation results of the relationship between the mode size and the width of the PE region, using film thickness as the parameter. There was a minimal value for the mode size at specific widths. For example, if the LN thin film thickness was 0.6 μm, the minimum mode size was reached when the PE region width was around 0.8 μm. As the waveguide width increased, the PE region expanded, and so the mode size became larger. When the width decreased, the confinement of light would become weak, which also led to a larger mode size. In the inset of Fig. 2, a 0.6 μm thick and 0.8 μm wide channel waveguide had a mode size as small as 0.6 μm² (product of 1/e intensity in horizontal and vertical directions). This value was comparable to the calculated mode size of a 1 μm wide ridge waveguide (etched LN ridge on SiO₂) reported in [15] (~0.4 μm²). The smallest mode sizes in etched LNOI waveguide were calculated to be as small as 0.15 μm² (0.35 μm wide ridge, TM polarization) and 0.16 μm² (0.45 μm wide ridge, TE polarization). The larger mode size in PE LNOI compared with ridge waveguide can be attributed to the weak confinement of light in the horizontal direction. However, the mode size we obtained in PE LNOI waveguide (0.6 μm²) was more than one order of magnitude smaller than a previously reported single-mode PE waveguide in bulk LN [30]. Small mode size means tight confinement of light, which benefits a stronger non-linear optical process. For channel waveguides in LNOI, light was strongly confined due to the large refractive index contrast between the LN layer and the SiO₂ cladding, which resulted in a small mode size. In bulk material, the guided mode existed only when its effective refractive index was larger than the refractive index of bulk material (about 2.138). However, In LNOI, even without PE, the LN layer formed a planar...
waveguide. The channel waveguide mode could be excited with an effective refractive index larger than that of the LN planar waveguide (e.g. about 1.78 at the thickness of 0.5 μm, which is much smaller than 2.138). The effective index would decrease with the shrinking of the waveguide dimension. Therefore, the effective index and the dimension of channel waveguide in LNOI had more of a chance to decrease, which meant it reached a more compact size.

![Graph](image1.png)

**Fig. 2.** Relationship between the calculated mode size and width of the PE region, using film thickness as the parameter (Δn = 0.09). Inset: A mode size as small as 0.6 μm² was obtained with a 0.6 μm thick film and a 0.8 μm wide PE region.

### 3. Experiment and results

Two identical Z-cut LNOI samples (#1 and #2) were prepared by crystal ion slicing and wafer bonding technology, in the research center of Nanoln, based on a previously described process [1]. The LN film was 500 nm thick and the structure of the sample is shown in Fig. 3. Sample #1 was used to investigate the refractive index change and phase transition in the LN thin film processed by the PE and sample #2 was used to fabricate channel waveguides.

![Diagram](image2.png)

**Fig. 3.** Schematic diagram of the LNOI cross section. h₁, h₂ and h₃ were 500 nm, 2 μm and 0.5 mm, respectively.

#### 3.1 Sample #1

Sample #1 was prepared under the following experimental conditions: Pyrophosphoric acid was applied as the proton source to exchange Li⁺ in LN. The PE was performed in a sealed furnace at 200 °C for 80 min, which was sufficient to make protons penetrate the whole LN layer [28], so the thickness of the PE region was the same as LN layer (0.5 μm). When the proton exchange duration was reached, the sample was hung in the furnace until the temperature gradually dropped to room temperature. Finally, the sample was cleaned with de-ionized water. The dark modes of sample #1, before and after the PE process, were measured using a prism coupler (Model 2010 Metricon, USA) at 633 nm and 1539 nm.
respectively. The measured effective refractive indices of the planner guiding modes at 633 nm are listed in Table 1. All of the effective extraordinary refractive indices of the LN planar waveguide modes increased after PE. The $n_e$ of the LN layer before and after PE, were 2.2018 and 2.3508 respectively as calculated from the effective refractive index of such modes. An increase of 0.149 was obtained at 633 nm and this value was 0.09 at 1539 nm. The film thickness, which was near the same before and after PE, was measured to be about 500 nm.

Table 1. Effective refractive indices of guiding modes in LNOI before and after PE at a 633 nm wavelength

| Mode   | Before PE | Mode 1 | Mode 2 |
|--------|-----------|--------|--------|
| Mode 0 | 2.128414  | 1.898200 | 1.535591 |
| Mode 1 | 2.268032  | 2.008343 | 1.580884 |

Figure 4 shows the X-ray diffraction (XRD) pattern around the LN (006) reflection for sample #1. The discrepancy between the LN substrate (39.22°) and LN film peak (39.32°) arose from the small deviation in the crystallographic Z-axis of the film from that of the substrate. After PE, a new phase occurred at the position of 39.1°. According to a previous report examining PE in bulk LN material [31,32], the new phase was $\beta_4$ phase ($\Delta n_e > 0.14$).

3.2 Sample #2

Sample #2 was used to fabricate the channel waveguides. A 100 nm thick chromium (Cr) layer was deposited on the surface of the sample via the electron beam evaporation method. A photolithography process was used to define the PE mask, which had open strips of 1 to 7 μm. After the exposed Cr was wet etched and the photoresist was removed by acetone, the PE was performed under the same condition as sample #1, so the thickness of the PE region was 500 nm. Finally, the remaining Cr mask was removed by Cr etchant ($NH_4)_2Ce(NO_3)_6 + C_2H_4O_2 + H_2O$ and the two facets of the fabricated waveguides were polished to facilitate the end face coupling. The final length of the sample was about 1.6 mm. Figure 5 shows the top view of the channel waveguide, which was defined by the 3 μm wide open mask. In this study we spent a long time to completely wet etch the exposed Cr. As a consequence, the width of the waveguide expanded to about 7 μm (increased by 4 μm) due to the lateral erosion of Cr in the wet etching process. This issue can be avoided in the future if another Cr deposited sample (deposited in the same condition) is used to calibrate the wet etching time to avoid excessive lateral erosion. In the following paragraphs, the width of the waveguides we mentioned are the value we observed by microscope rather than the opened mask width.
A tunable semiconductor laser (Santec TSL-210) was used as the near-infrared light source (1260 nm-1630 nm). The linear polarized light emitted from this laser was transmitted through a polarization-maintaining fiber and then rotated to a TM polarized direction by a rotator. The light was coupled into the waveguide by the lensed tip of the fiber and the output light was collected by a 40 × / 0.65 objective lens. A knife-edge based beam profiler (BM-7, Coherent Inc.) and germanium photodiode were used to record the near-field intensity profile and the power of light, respectively. As shown in Fig. 6, the measured mode sizes (1/e intensity in horizontal and vertical directions) were about 3 μm × 1.1 μm (3.3 μm²), 4.5 μm × 1.1 μm (5 μm²) and 6.4 μm × 1.1 μm (7 μm²) and the theoretical values were 3.6 μm × 0.38 μm (1.4 μm²), 4.7 μm × 0.38 μm (1.8 μm²) and 7 μm × 0.38 μm (2.7 μm²), respectively. Considering that the vertical dimension of the waveguide (0.5 μm) is smaller than the resolution of the objective (~1.5 μm), the real profile and dimension in the vertical direction cannot be completely reflected, leading to the larger mode sizes (three times the theoretical value in vertical direction). Other optical characterization techniques, such as Near-field Scanning Optical Microscopy, which is used to observe sub-micron scale photonic structures, is suitable if the local characteristics of guided modes in sub-micron scale photonic structures is needed [33]. The measured dimension in horizontal direction was a little smaller than the simulated one. This probably indicates a difference between the model we used in simulation and the actual structure of PE LNOI waveguide: the step-like index profile used in simulation is an approximate assumption; and in simulation, the refractive index change in channel waveguide was assumed to be 0.09, which was the measured value in planar waveguide. However, even under the same PE condition, the refractive index change in channel waveguide and in planar waveguide are not exactly the same.

Fig. 6. Measured near-field intensity distributions of the fundamental quasi-TM modes, guided in the (a) 5, (b) 7 and (c) 11 μm wide channel waveguides at 1.55 μm. The corresponding simulated waveguides are shown in (d), (e) and (f). Mode sizes of 3.3 μm², 5 μm² and 7 μm² were obtained by measurement and mode sizes of 1.4 μm², 1.8 μm² and 2.7 μm² were obtained by simulation.
The Fabry-Perot resonator method was used to evaluate the propagation loss $\alpha$ of the channel waveguides. The polished end-faces of the waveguide were regarded as two Fabry-Perot resonant mirrors. The propagation loss was calculated through the transmission spectrum, which is shown in Fig. 7. The oscillating curve was due to Fabry-Perot interference and the mode propagation loss was determined by analyzing the contrast $K$ of the cavity resonances [34]. The formulas to evaluate $\alpha$ and the related parameters were defined as follows:

$$\alpha = \frac{4.34}{L} (\ln R - \ln \tilde{R}) \quad \text{where} \quad \tilde{R} = \frac{1}{K} (1 - \sqrt{1 - K^2})$$

and

$$K = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \quad (1)$$

$I_{\text{max}}$ and $I_{\text{min}}$ were the maximum and minimum intensity of the transmitted light, which were extracted from Fig. 7. The modal reflectivity $R$ of the 5, 7 and 11 $\mu$m wide channel waveguide end face, calculated by a finite difference time domain (FDTD) solver, were 0.107, 0.106 and 0.105, respectively [26,27]. The loss $\alpha$ was evaluated to be about 16, 12 and 11 dB/cm. These values were comparable to the dry etched ridge waveguide in LN thin film (around 10 dB/cm at 1.55 $\mu$m) [15] and were larger than the wet etched ridge waveguide (1.3 dB/cm at 0.6328 $\mu$m) [35] and the PE waveguide in bulk LN (1 dB/cm at 0.78 $\mu$m) [30]. More efforts should be made to decrease the propagation loss. The loss came from several sources. First of all, a study has shown a degradation of the device performance (extinction ratio, $V_\pi L$ and insertion loss) due to the helium-ion-induced damage during the LN thin film preparation [36]. The degree of the overlap between the ion-damaged region and the guided optical modes has a significant effect on the degree of achievable modulation. Therefore, the helium-ion-induced damage in the PE region in our work is regarded to be a possible reason of the propagation loss. Secondly, LN layer lattice structure and constituent have changed from the PE process. The pure $\alpha$ phase waveguide has lower propagation loss than the $\beta$ phase and the coexistence of two different crystal phase because the $\alpha$ phase has the least changes in lattice constant and the refractive index [37]. Next, the scattering loss might occur from the interface of the LN film and SiO$_2$ layer. Finally, for multi-mode waveguide, nearly always a combination of modes is excited by end-face coupling. This will lead to a reduction of the measured contrast $K$ and an increased loss. Although the intensity distribution illustrated a fundamental mode profile, some higher order modes might be partly excited. In addition, imperfections induced by polishing would lower the reflectivity of the waveguide end face. So the loss number we obtained from formula (1) is the upper limit for the fundamental mode [33].

Therefore, to improve the quality of the PE waveguide, some parameters including implanted energy and dose and the following annealing condition in the process of LNOI fabrication, should be controlled and optimized to get the least helium-ion-induced damage in the LN thin film. The $\alpha$ phase waveguide should be fabricated since it changes the LN crystal structure the least among all of the PE LN phases. This could be done by performing PE in benzoic acid for a short time, followed by annealing or immersion in a benzoic acid melt, buffered with lithium benzoate for a long time [32,38]. To accurately evaluate the propagation loss, we need to fabricate single mode waveguide to avoid the reduction of the measured contrast $K$. 
4. Conclusions

In conclusion, PE waveguides in LNOI were studied. Simulations, including single mode conditions and mode size calculations, were performed. Compared with previously reported PE channel waveguides in bulk LN, a much smaller (more than one order of magnitude) mode size in LNOI, of about 0.6 μm², was obtained. The small mode would enhance the nonlinear and electro-optical effect, leading to high efficiency all-optical signal processing and E-O modulation. The simulation also showed that, in the β₄ phase, the channel waveguide was single mode when the PE width was between 0.75 μm and 2.1 μm. The dark-modes before and after PE were measured, for the planar waveguide, and an increase in nₑ of 0.149 at 633 nm, was obtained. X-ray diffraction indicated a new phase (β₄) emerging in the PE LNOI. The fabrication and optical characterization of the PE channel waveguide on LNOI were presented. A standard photolithography followed by a PE process was performed on a LNOI sample with a 0.5 μm thick LN layer. For the 5 μm, 7 μm and 11 μm wide channel waveguides, the near-field intensity distribution showed 3.3 μm², 5 μm² and 7 μm² mode sizes, and the propagation losses of them, which were obtained using the low-finesse Fabry-Perot resonator method, were 16 dB/cm, 12 dB/cm and 11 dB/cm, respectively. The fabrication of a low loss, single mode, α phase PE waveguide and electro-optical modulator in LNOI are now in progress. The simulation and successful fabrication of a PE LNOI waveguide confirmed that these two mature techniques (PE and LNOI fabrication) are compatible. This could lead to the realization and use of more advanced and complicated photonic integrated devices and circuits, based on PE LNOI, in the future.

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