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PROTON-EXCHANGED $LiNbO_3$ WAVEGUIDES: MATERIALS ANALYSIS AND OPTICAL CHARACTERISTICS

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Proton-exchanged $LiNbO_3$ waveguides: materials analysis and optical characteristics

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Abstract

This paper reports on the relationship between materials analysis studies and optical characterisation of proton-exchanged lithium niobate waveguides. Close agreement in predicted waveguide depths is found on Z-cut $LiNbO_3$ substrates, while a consistent discrepancy is found on X-cut $LiNbO_3$ substrates.

Introduction

The emergence of the proton-exchange technique for optical waveguide fabrication[1] could have a major impact on the use of lithium niobate in various applications in integrated optics. Several interesting device possibilities have already been demonstrated[2-5] and some important practical problems have been identified[3,5,6]. There is clearly a need to establish unambiguously the optical characteristics of waveguides formed by the proton-exchange process to give a basis for the design of well-controlled and reliable devices and sub-systems. Several papers[7-9] have reported that optical measurements indicate that waveguides formed by proton-exchange have step-index profiles, but independent verification is clearly desirable.

This paper reports on the relationship between materials analysis studies and optical characterization of proton-exchanged lithium niobate waveguides fabricated in Z- and X-cut substrates immersed in benzoic acid at temperatures ranging from 150°C to 220°C and for times ranging from twenty minutes to a few hours.

Optical characteristics have been obtained primarily from an analysis of mode-angle measurements using the standard prism-coupler technique. Proton concentration and depth profile have been estimated via the nuclear reaction $H^1(N^{15}, \alpha\,\gamma)C^{12}$, while use of Rutherford back-scattering (RBS) in the 'aligned' and 'random' configurations has been used to identify the region where the proton-exchange process produces a pronounced lattice modification. Stresses and crystal structure modification have also been detected using double-crystal X-ray diffraction.

Waveguide sample fabrication

Single-crystal lithium niobate substrates were cleaned and degreased thoroughly using a series of organic solvents. The samples were then mounted in a PTFE holder before being placed in 250ml of benzoic acid in a stainless-steel beaker placed in a high-temperature oil-bath controlled to within ± 0.25°C. Use of a stainless steel beaker ensured good thermal contact between the acid and the high-temperature oil, while the volume of acid was large enough to provide an effectively infinite source diffusion. The benzoic acid was renewed after, at the most, twenty exchange runs. Within experimental accuracy, both optical waveguide measurements and IR absorption spectra showed no difference between samples fabricated in fresh benzoic acid and samples from the twentieth run. Both oil-bath and exchange-beaker were covered to provide a well-isolated temperature-stable environment.

Exchange-temperatures used ranged from 150°C to 220°C with sample pre-heating being required at the lowest temperatures. The exchange time was defined as that between immersion of the sample in the benzoic acid and its removal from the acid. Crystallized benzoic acid, which formed on the $LiNbO_3$ substrate because of cooling after exchange, was removed by rinsing in chloroform and/or methanol. The samples were then stored for a long enough period for the refractive index of the waveguide region to relax to a stable value[6] (typically for at least two weeks).

Optical characterisation

The mode-structure of the optical waveguides was measured by means of the conventional prism-coupler technique with single-crystal rutile prisms. Analysis of output-mode coupling-angle data was based on the paper by Kogelnik and Ramaswamy[10]. (See also the earlier work of Snyder and De La Rue[11].) An initial assumption was made that data obtained corresponded to a step-index slab waveguide model. The appropriateness of this model was then verified by obtaining consistent results for estimated waveguide depth over a wide range of fabrication conditions and at two different wavelengths (0.633μm and 1.15μm).

For TE modes a normalized guide effective index b is defined:

$$b = \frac{(N^2 - n_f^2)}{(n_f^2 - n_s^2)} \tag{1}$$

where N is the guided-mode effective index (determined by measuring prism-coupler m-line angles), n_s is the substrate refractive index (known), and n_f is the (unknown) guiding-region refractive index. A normalised guide thickness V is defined:

$$V = kf(n_f^2 - n_s^2)^{\frac{1}{2}} \tag{2}$$

where f is the guiding region thickness and k is the free-space propagation constant. Plots of b versus V are obtained (Kogelnik and Ramaswamy Fig. 2) from solutions of the normalised dispersion equation (Eq. 9 of the same reference) with an asymmetry parameter $\underline{a}$ given by:

$$\underline{a} = \frac{(n_s^2 - n_c^2)}{(n_f^2 - n_s^2)} \tag{3}$$

where n_c is the cover region refractive index.

From a knowledge of the number of modes which the guide supports, an appropriately restricted range of values for V is determined and within this range a computer scan is carried out to obtain an estimate of n_f for each mode, given an initial estimate of the asymmetry parameter $\underline{a}$. b-values from an initial choice of V are obtained from the theoretical plots, and, together with N-values from measurement, are used to obtain sets of estimated n_f values. V is then adjusted (and consequently the set of b-values) until the standard deviation over a set of n_f-values is reasonably small (typically 10^{-4} for guides supporting several modes). The value for the asymmetry parameter $\underline{a}$ given by Eq. 3 is also refined at the same time. Having obtained an estimate of n_f and its corresponding value of V, the waveguide depth, f, then follows immediately from Eq. (2).

Strong support for the step index model stems from the fact that the same estimates of depth (to within 1%) are obtained over a range of waveguides at both 0.633μm wavelength and 1.15μm wavelength, with respective values, on X-cut $LiNbO_3$, of $\Delta n = 0.1267$ and $\Delta n = 0.096$. The TE-mode analysis described above applies for proton-exchanged guides on X-cut lithium niobate, while for the TM modes which occur on Z-cut lithium niobate, some modification of the theory is required[10].

Experimental estimates of propagation loss in our waveguides have been reported elsewhere[7,8].

Exchanged-region depth-measurements by RBS

In Rutherford backscattering spectrometry (RBS) a beam of $^4He^+$ ions in the 1.5 to 2.5MeV range bombards the sample surface. A small fraction of the incoming particles are backscattered and detected after Rutherford collisions with sample atoms. Energy analysis of the primary ions backscattered from the sample provides a mass-analysis of the atoms near the surface[12,13]. The relative sensitivity of different atoms is determined by the Rutherford scattering cross-section (proportional to Z^2, where Z is the atomic number) and this permits a "first principles" determination of concentration. Due to energy loss suffered by the probing particles as they traverse successively deeper atomic layers, energy analysis of the backscattered particles also provides in-depth distributional information.

When ions are incident on a single crystal within $\approx$ 1% of a high symmetry axis, they can be steered down an open channel in the lattice by the repulsive potential of atomic strings, as shown in Fig. 1(a). This effect, called channelling, reduces the probability of nuclear scattering by as much as two orders of magnitude. In such channeling conditions, surface damage due, for example, to the mechanical polishing and epitaxial nature of surface layers can easily be studied.

Fig. 1(c) shows, schematically, backscattering spectra in channelling conditions for a perfect $LiNbO_3$ crystal and for a $LiNbO_3$ crystal substrate with a surface layer whose atom positions are distorted with respect to those of a perfect crystal owing to the proton-exchange process. In this latter case the backscattering spectrum shows an increase in the backscattering yield for an energy range ΔE which is proportional to the thickness W of the exchanged (distorted) layer.

Fig. 2 shows the experimental RBS spectra for Z-cut $LiNbO_3$ substrates exchanged in pure benzoic acid for 1 hour at four different temperatures. The aligned spectra clearly show a surface region with a slightly higher backscattering yield with respect to the virgin sample owing to a small displacement of atoms with respect to their regular positions in the perfect crystal lattice. The depth of the proton-exchanged and, therefore distorted layer increases with increasing temperature at the fixed exchange time of 1 hour. Furthermore, for all samples the small peak present at the surface suggests a small amount of surface damage essentially due to mechano-chemical polishing of the samples. Similar results have been obtained on X-cut substrates.

The exchanged region depths measured in this way agree very well with those measured by guided optical techniques in the case of Z-cut substrates, while optical results show a value consistently larger by about 30% for X-cut samples.

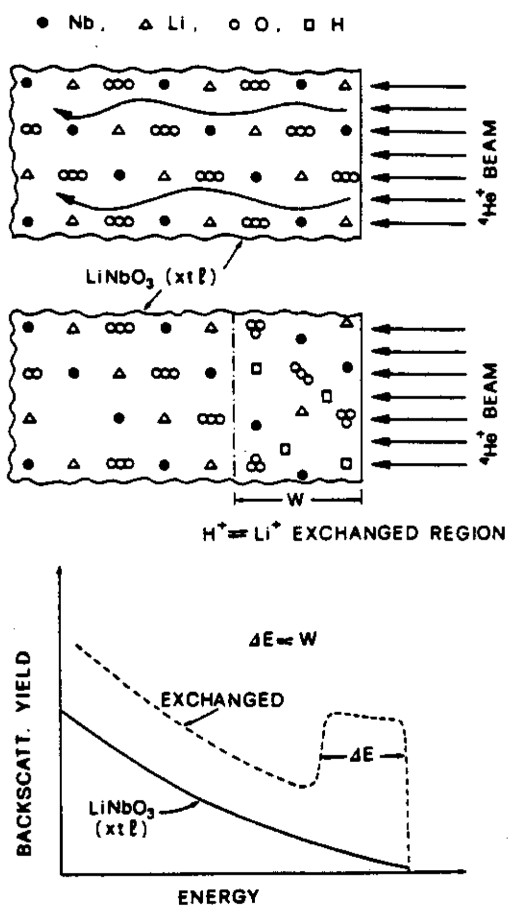


Fig. 1. Schematic RBS spectra taken in aligned conditions for a perfect $LiNbO_3$ crystal and for one with a distorted surface layer due to the proton-exchange process

Hydrogen profiling by nuclear reaction (N.R.)

A complementary nuclear technique, especially useful for the analysis of elements of low atomic number (where RBS loses sensitivity owing to the Z^2 dependence of the cross-section) is based on the analysis of nuclear reaction products induced by the bombarding charged particles. NR can provide isotope-sensitive information[14]. The observed reaction products are usually charged particles which are detected and analysed by means of nuclear detectors. In NR the choice of the projectile, its energy, the element to be analysed, angle of detection and detector arrangement must all be carefully considered.

When the differential cross section for nuclear reaction is nearly constant as a function of projectile energy, the nuclear reaction which occurs is particularly suitable for determining the total number of investigated nuclei. However, a resonance in the cross-section can be most useful for depth profiling.

In particular the N^{15} hydrogen profiling technique uses a narrow isolated resonance in the $N^{15} + H^1 \rightarrow C^{12} + H_e^4 + 4.43MeV$ γ-ray process to measure the hydrogen content versus depth[15]. Furthermore the off-resonance cross-section of this nuclear reaction is three orders of magnitude smaller than the peak value. To use this resonance as a probe for hydrogen, the sample is bombarded with N^{15} and the yield of the characteristic 4.43MeV γ-rays is measured with a NaI detector. When the sample is bombarded with a N^{15} beam at the resonant energy, $E_R = 6.385MeV$, the yield of characteristic γ-rays is proportional to the hydrogen content on the surface of the target. When the beam energy is raised above the resonant energy, surface hydrogen is no longer detected because the N^{15} is above the resonant energy, but, as the N^{15} beam slows down in the energy while passing through the target it reaches the resonant energy at some depth. The γ-ray yield is now proportional to hydrogen concentration at this depth, which can be estimated accurately because of the known energy loss behaviour for ions in solids. Hence, by measuring γ-ray yield versus the energy of the incident N^{15} beam, the concentration of hydrogen vs. depth in the target is determined. The interaction between N^{15} beam and the target discussed above is shown schematically in Fig. 3. The depth resolution of this method is finite because the resonance has a finite energy width, ± 3KeV, which corresponds for $LiNbO_3$ to a depth resolution of approximately 20Å. The sensitivity of this method for H detection is about 0.1 at %.

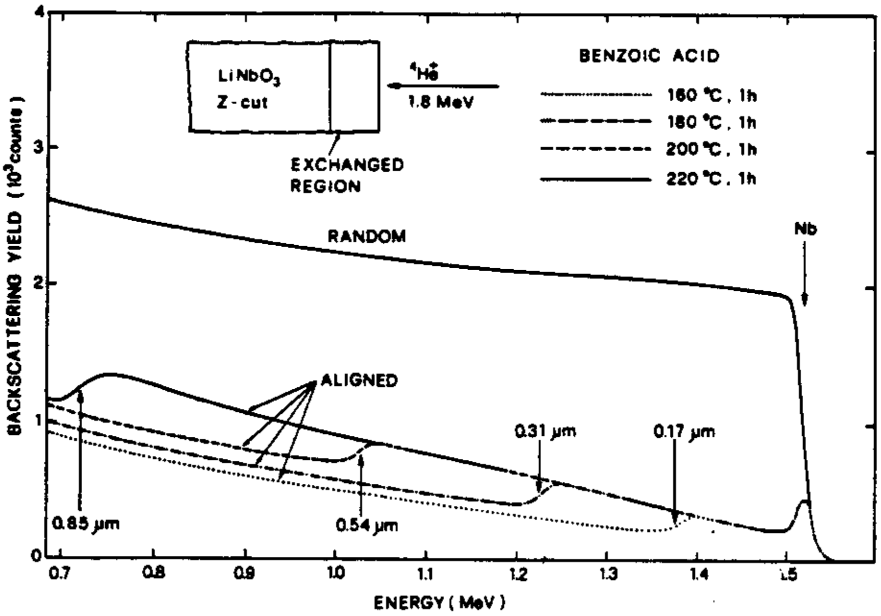


Fig. 2. RBS spectra taken in aligned conditions on Z-cut samples exchanged in pure benzoic acid at four different temperatures for 1 hour. The Nb arrow indicates the energy of $^4H_e^+$ particles backscattered from Nb atoms on the sample front surface

Fig. 4 shows the measured hydrogen profiles in Z- and X-cut LiNbO$_3$ samples exchanged in pure benzoic acid at two temperatures, 160 and 180°C for several different times.

The data clearly show a step-like hydrogen distribution with a well defined plateau where the resultant H-concentration is in the range 1.1 to 1.3 10^{22} cm^{-3} with apparently, a small dependence on exchange temperature and substrate crystallographic orientation. The concentration is slightly higher for X-cut material. The depth of the exchanged region measured by nuclear reaction always agrees closely with RBS data, but for optical measurements again good agreement was found for Z-cut samples while for the X-cut samples, the NR results are lower by about 30%.

Discussions and conclusions

Optical waveguides on Z- and X-cut LiNbO$_3$ single crystal substrates, proton-exchanged in pure benzoic acid at different temperatures and times, have been optically characterised and structurally analysed. Optical waveguide measurements at both $\lambda = 0.6328$ and at $\lambda = 1.15\mu m$ clearly support a step-index model for the waveguide profile. Structural analyses were mainly performed using high energy ion beams. RBS data taken in channelling conditions reveal the presence of slightly distorted lattice conditions in the exchanged zone, thus allowing the measurements of the exchange-region depths as a function of exchange temperature and time. The increase of backscattering yield in the aligned spectra corresponding to the exchanged zone may be due to a distortion of the lattice parameters and atom locations due either to a H → Li substitution or to the possible presence of interstitial H atoms in the crystal structure.

The presence of strains in the exchanged layer was also detected by the double crystal X-ray rocking curve technique. In fact, in the exchanged samples, Fig. 5, the (220) rocking curves were accompanied by various diffraction satellite peaks with both positive and negative angular shifts; whose interpretation clearly requires a quite complicated computer simulation.

The $H^1(N^{15}, \alpha\gamma)C^{12}$ nuclear reaction can provide directly the total hydrogen concentration and in-depth profiles which show a typical step-like form. In the constant concentration zone the measured H-content is in the region of 1.1 to 1.3 x 10^{22} cm^{-3} and corresponds to H/Li ratios in the range 0.58 to 0.72 (assuming that the number of Li atoms present in LiNbO$_3$ is 1.88 x 10^{22} cm^{-3}) in broad agreement with data reported by Jackel et al[16].

Measurements of the exchanged region depths obtained with nuclear reaction and RBS in

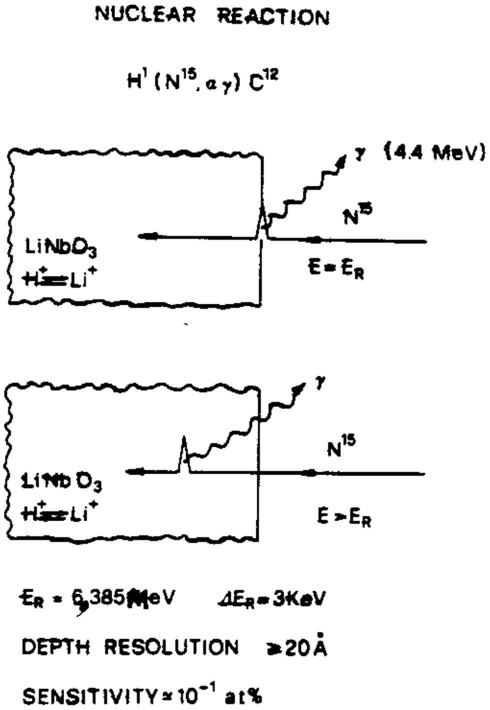
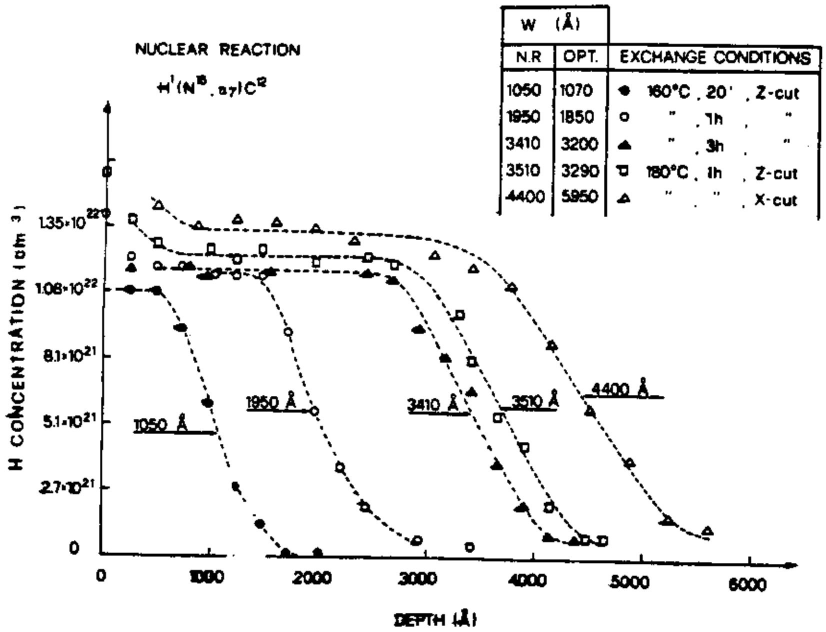


Fig. 3. Schematic of the N^{15} resonance profiling method. The N^{15} beam bombards the ~~target~~ a) with $E = E_R$ and b) with $E > E_R$. The number of γ-rays is proportional to hydrogen concentration a) at the surface and b) at the depth where N^{15} beam slows down to $E = E_R$ passing through the target.



W (Å)		
N.R	OPT.	EXCHANGE CONDITIONS
1050	1070	● 160°C , 20' , Z-cut
1950	1850	○ " , 1h , "
3410	3200	▲ " , 3h , "
3510	3290	□ 180°C , 1h , Z-cut
4400	5950	△ " , " , X-cut

Fig. 4. Measured hydrogen profiles on $LiNbO_3$ Z- and X-cut samples exchanged in pure benzoic acid at two temperatures, 160 and 180°C, and different times

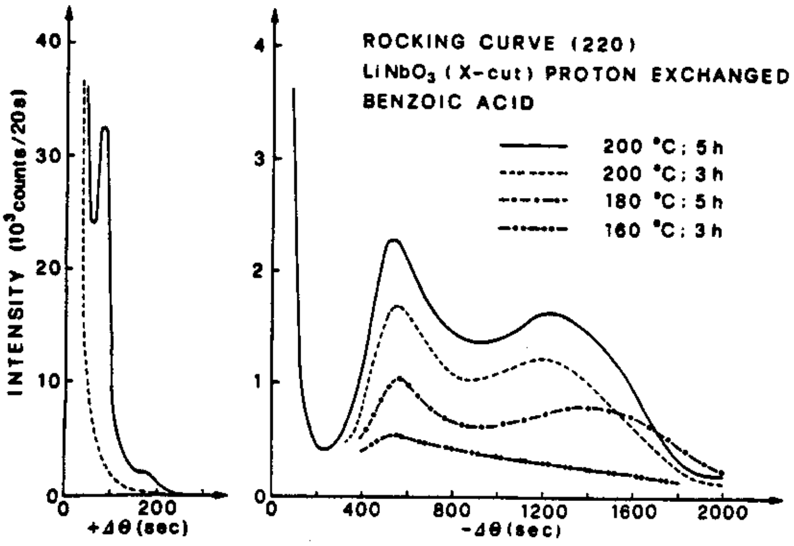


Fig. 5. Double crystal X-ray diffraction (220) rocking curves obtained on X-cut samples exchanged at different times and temperatures in pure benzoic acid

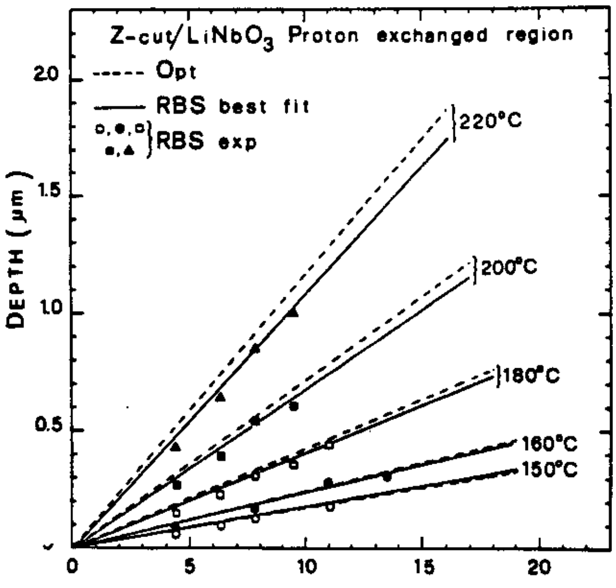


Fig. 6. Depths of exchanged regions measured by RBS in channelling condition and with optical techniques on Z-cut LiNbO$_3$ samples exchanged at different temperature and times in pure benzoic acid.

channelling conditions agree well with each other, thus proving that distortions of lattice parameters and/or of atomic locations are strongly correlated to the presence of protons. On Z-cut substrates, there is good agreement between depth estimates obtained from optical waveguide measurements[7,8] and measurements of exchanged layer depths obtained using ion-beam techniques as is shown in Fig. 6. For X-cut substrates, optical depth estimates appear to be about 30% lower than ion-beam depth measurements. We are not able to give a clear explanation for this disagreement at this time. However, we have shown that a close correlation between optical and structural properties, in particular proton atomic concentration profiles and defects induced by the exchange process, is possible and we believe that this will help in understanding the proton-exchange process in $LiNbO_3$ and therefore lead to improved optical waveguide performance and reliability.

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