

Proton exchange technology for optical waveguides in lithium niobate and lithium tantalate – an overview

M. Kuneva*

Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Blvd, 1784 Sofia, Bulgaria

Received October 10, 2016; Revised November 20, 2016

Lithium niobate (LN) and lithium tantalate (LT) are nonlinear, birefringent crystals with an important role in optoelectronics. Beside their excellent optical properties, their most prominent feature is the high electro-optical coefficient ($r_{33} \approx 30.5$ pm/V) which allows the light field to be easily controlled by electric signals. Proton exchange (PE) is a method of obtaining $\text{LiM}_{1-x}\text{H}_x\text{O}_3$ layers ($M = \text{Nb, Ta}$) on LiMO_3 substrates. The PE layers have a significantly higher extraordinary refractive index (n_e) than the non-protonated crystal; the change is $\Delta n_e = 0.120\text{--}0.150$ for LN and $\Delta n_e = 0.015\text{--}0.020$ for LT at wavelength $0.633 \mu\text{m}$ providing a strong waveguide and polarizing effect.

The present paper aims at summarizing the most significant technological modifications of the method of PE and the most important waveguide structures of $\text{LiM}_{1-x}\text{H}_x\text{O}_3$ for different modulators used in modern optoelectronic devices (navigation and communication systems, biosensors, etc.).

Keywords: proton exchange, optical waveguides, integrated optics

INTRODUCTION

Two of the most widely used ferroelectric materials in integrated optics (IO) are lithium niobate - LiNbO_3 (LN) and lithium tantalate - LiTaO_3 (LT). This is due to their high electro-optical (EO) coefficients ($r_{33} \approx 30.5$ pm/V) and the possibility to change the refractive index by modifying their composition in regions where waveguiding of light is required. These features, combined with excellent optical properties and suitability for industrial production, make them key materials for photonics.

Proton exchange is a technology with already well-studied advantages and known prospects for the obtaining of optical waveguides in LN and LT, as well as of a wide range of passive elements and active optoelectronic devices.

PROTON EXCHANGE TECHNOLOGY

Proton exchange is an induced - at favorable conditions - diffusion of protons into the surface layer of a crystal substrate, whereby the layer's refractive index becomes higher than that of the substrate. The method is interesting with its simplicity and fastness, combined with the possibility of a strong waveguide effect (a

significant change of the refractive index), low diffusion temperature ($150\text{--}300$ °C, of particular importance for LT) and decreased photorefractive susceptibility. Proton exchange increases the extraordinary refractive index (up to 0.15 for LN and 0.02 for LT at 633 nm) and lowers the ordinary refractive index; therefore, depending on the orientation of the substrate, the propagation of only one polarization is sustained (TE for X- and Y-cut samples or TM for Z-cut samples), i.e. PE waveguides guide only light polarized along the optical axis of the crystal. Going by the scheme:



PE modifies the surface layer (several μm in depth) by Li-H ion exchange at a relatively low temperature ($160\text{--}250$ °C), usually in acidic melts. The diffusion is anisotropic and the value of the diffusion coefficient depends on the substrate crystallographic orientation. This process changes the structure and the composition of the exchanged area. The PE layers show complex phase behavior depending on the hydrogen concentration (the value of x). The value of x determines the concentration limits of the different phases that could form in the waveguide layer (up to 7 in LN and 5 in LT) [1, 2]. The formation of phases depends in a complex way on the crystallographic orientation and the diffusion parameters. Each phase forms a separate sublayer of submicron thickness, and differs from the others by its structural and optical properties. The phases

* To whom all correspondence should be sent:
E-mail: m_kuneva@yahoo.com

also have different lattice parameters. At the interface of two phases, a rapid change of the extraordinary refractive index (Δn_e) and the deformation perpendicular to the surface is observed. Within each phase, Δn_e is proportional to x . The phase composition determines the properties and the quality of the obtained waveguides. Strong protonation considerably worsens the electro-optical properties, causes higher losses and some instability of the parameters over time. These can be avoided by using the methods given below for optimization of proton concentration and optical profiles of the PE layers:

PE – proton exchange [3] – a one-step process which takes place when LN or LT substrate is immersed in an appropriate proton source. PE waveguides usually have step-like optical profile and nearly vanished EO-coefficients [4].

DPE – deep proton exchange [5] – proton exchange for achieving a large change of the refractive index ($\Delta n_e=0.15$). Optical profile is almost step-like, better approximated by two rectangular steps (or truncated parabolic profile shape) [6].

ADPE – DPE with subsequent long time and high temperature annealing (two-step process) applied for obtaining deeper (over 20 μm thick) waveguides [5].

VPE – PE in vapors (one-step process) [7, 8]. VPE allows highly homogeneous monophasic waveguides with very low propagation losses to be obtained by PE and reverse diffusion within a single chemical reaction [7].

SPE – soft proton exchange in buffered melts (i.e. lithium benzoate added to the benzoic acid) - one-step process for obtaining of waveguides with preserved EO properties [9].

HTPE – high temperature PE (one-step process). Acid with a higher boiling point and lower vapor pressure is used as a proton source [10]. High quality α -phase waveguides could be obtained directly (with no phase transitions) for a short time.

APE – PE with subsequent annealing (two-step process) for obtaining monophasic (α) waveguides with restored EO properties after phase transitions from phases of high values of x to the phase of lower value of x [11].

PEAPE – two-step PE with intermittent annealing (three-step process) [12, 13], appropriate for obtaining deep waveguides with high refractive index change in Y-cut LN.

RPE – reverse PE (two-step process) [14] for obtaining buried waveguides with various

properties. Takes place when PE-waveguide is immersed in eutectic mixture of LiNO_3 , KNO_3 and NaNO_3 .

RAPE – RPE in APE waveguides (three-step process) [15] used to bury the waveguide under the crystal surface, increasing that way the circular symmetry of the optical mode.

Thus a predetermined phase composition that meets particular requirements – a high refractive index, high electro-optical coefficients, low optical losses, stable parameters over time, decreased photorefractive susceptibility, or combinations thereof – can be achieved with an appropriate selection of proton exchange conditions.

APPLICATIONS IN PHOTONICS, OPTOELECTRONICS AND SENSORS

Proton exchange is mainly used for the obtaining of high-quality waveguides for different modulators, switches, multiplexers and Y-splitters as main elements of modern optoelectronic devices for navigation equipment, communication systems, in a number of sensors (detectors of molecules in fluids, biosensors, contamination detectors, temperature sensors, etc.) and many devices for interferometric control. Proton-exchanged LN and LT waveguide devices are favored over Ti-diffused LN ones in cases where high optical powers are to be transmitted and/or single polarization operation is desired. Some examples illustrating the use of the PE technology in modern optoelectronics are given below.

1. Phase modulator

Phase modulation originates from the electro-optical properties of LN (LT) when the refractive index changes, causing phase variation of propagating light. Principle schemes of three modifications of the phase modulator are shown in Fig.1. The device consists of a stripe waveguide (2) formed in the crystal substrate (1) and electrode configuration (3) (Fig. 1 a and b). When electric field is applied via electrodes, a phase change ($\Delta\phi$) takes place in the light travelling along the electrodes (longitude L). The phase shifting is expressed by

$$\Delta\phi = k\Delta nL \quad (1)$$

where k is a wave factor $k=2\pi/\lambda$ (λ is the light wavelength in vacuum) and Δn is the refractive index change.

As a phase shifter, this modulator is an important component in many EO lightguiding

structures, due to its simplicity, efficiency and high speed, e.g., in different interferometric systems for phase deviation or compensation in the two arms. The EO phase shifters are very popular in fiber optic sensor technology because of the advantage that the phase measurements are not influenced by amplitude variations.

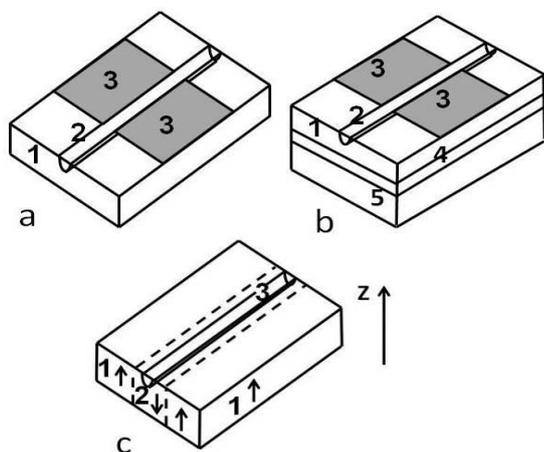


Fig. 1. A principle scheme of a phase shifter (a); PELN on insulator phase modulator (b): 1 – substrate, 2 – PE stripe waveguide, 3- electrodes, 4 – SiO₂ layer; schematic representation of a dipole inverted PE element for electric field sensor (c): 1, 2 – DI regions, 3 – PE stripe

An example of the application of combined advanced technologies for obtaining a phase modulator is discussed in [16]. A proton-exchanged stripe waveguide (near cut-off) is formed in the LN on insulator film (Fig. 2b). The lithium niobate on insulator technology (LNOI) is attractive due to the high optical confinement and enhanced lightguiding capability. The structure has a low driving voltage and could be used in high sensitivity sensors.

Another example of a device based on dipole inverted (DI) PELN is an all-optical electric field sensor proposed in [17]. A proton-exchanged stripe waveguide (3) near cut-off is fabricated in the central DI region (2) of a Z-cut LN substrate (Fig. 2c). When external electric field is applied parallel to the z-axis of the substrate, the refractive index in the DI region increases/decreases but that in the surrounding domains (1) decreases/increases causing mismatch of the guided modes between active and passive regions and thus produces a loss after a sufficient propagation length. Electric field intensities up to 20 MV/m could be measured this way.

2. Mach-Zehnder (MZM) modulator/interferometer

The Mach-Zehnder interferometer structure is

the most frequently used type of EO amplitude modulator and is a basic structure in a variety of IO devices. As in the classical bulk configuration of the MZ interferometer, light splits into two optical paths with no possibility of interaction. The key point in the interferometric modulators working on the Mach-Zehnder principle from classic optics consists of (in their integrated-optical version) two Y-junctions which perform the function of beam splitting and coupling into the stripe waveguides which represent the interferometer's channels (Fig. 2a). The refractive index (i.e. the propagation velocity) in each arm could be modified by applying electric field, changing that way the interferometric pattern at the output. The performance of the device strongly depends on the electrode configuration and waveguide architecture.

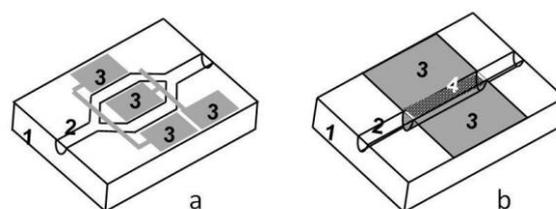


Fig. 2. Principle schemes of MZM: (a) - classical configuration; (b) – single-stripe MZM; 1 – substrate, 2- PE-stripe (supports single mode), 3 – electrodes, 4 – deeper stripe (supports 2 modes)

A fundamentally new, technologically simpler broadband IO modulator - a single-stripe MZM (Fig. 2b) has been designed based on the polarizing action of PELN [18]. The waveguide structure is formed in the LN X-cut wafer (1) and consists of a single-mode channel waveguide (2) whose central region (4) is deeper (produced via double proton exchange), has a higher rate of refractive index change and supports two modes. This new construction uses the simplest electrode configuration consisting of two parallel driving electrodes (3). Upon reaching region (4), light divides into two orthogonal modes which propagate independently, and after voltage is applied to the electrodes, the phase for each optical mode changes in a different way because the overlapping integrals for electric and optic fields are different for each mode. Reaching the end of the two-mode region, these two modes interfere and the light continues propagating as a single mode whose intensity depends on the phase difference between modes in the central region, i.e. on the voltage applied. This construction together with the technology used allows easier control of the geometry and the composition of waveguiding regions as well as

further device minimization (long Y-splitter's length being avoided).

CONCLUSIONS

Due to the importance of PELN and PELT for integrated optics, a variety of methods have been developed for fabrication of waveguides in these materials but none of them is universally applicable. Very promising appear to be the combined methods, and the efforts are aimed at the introduction of some technological modifications of the existing methods.

The PE technology has left its laboratory research level and reaches the manufacturing stage. Integrating single components and structures on a common chip allows a great variety of IO devices to be obtained.

The future developments will tend to attract new technologies like photonic crystal technologies and nanotechnologies for solving the problem of input and output coupling with application of IO devices. The achievement of higher data processing speeds and an increased level of integration are also among the current objectives of the development of PELN and PELT based optoelectronics.

REFERENCES

1. Yu. Korkishko, V. A. Fedorov, *Proc. SPIE*, **2700**, 186 (1996).
2. Yu. Korkishko, V. Fedorov, *IEEE J. Select. Top. Quant. El.*, **2**, 187 (1996).
3. J. L. Jackel, C. E. Rice, J. J. Veselka, *Appl. Phys. Lett.*, **41**, 607 (1982).
4. A. Méndez, G. De La Paliza, A. Garsia-Cabanes, J. M. Cabrera, *Appl. Phys. B*, **73**, 485 (2001).
5. M. Okazaki, T. Chichibi, S. Yoshimoto, H. Mizuna, and T. Suhara, *Jap. J. Appl. Phys.*, **54**, 301 (2015).
6. M. Kuneva, S. Tonchev, M. Sendova-Vasileva, D. Dimova-Malinovska, P. Atanasov, *Sensors and Actuators A*, **99**, 154 (2002).
7. M. Kuneva, S. Tonchev, P. Dimitrova, *J. Mat. Sci.:Materials in Electronics*, **14**, 859 (2003).
8. J. Rams, J. M. Cabrera, *J. Opt. Soc. Am. B*, **16**, 401 (1999).
9. K. Wong (1985). *Gec. J. Research*, **3**, 243 (1985).
10. Yu. N. Korkishko, V. A. Fedorov, O. Y. Feoktistova, *J. Lightwave Technol.*, **18**, 562(2000).
11. S. Al-Shukri, A. Dawar, R. De La Rue, A. Nutt., M. Taylor, J. Tobin, *Proc. 7th Top. Meet. On Integrated and guided wave optics* **PD7**, 1 (1984).
12. M. Kuneva, S. Tonchev, M. Pashtropanska, *Cryst. Res. Technol.*, **32**, 387 (1997).
13. M. Kuneva, S. Tonchev, M. Pashtropanska, I. Nedkov, *Materials Science in Semiconductor Processing*, **3**, 581 (2001).
14. Yu. N. Korkishko, V. A. Fedorov, T. M. Morozova, F. Caccavale, F. Gonella, F. Segato, *JOSA A*, **15**, 1838 (1998).
15. H. Hui, R. Richen, W. Sohler, Proc. ECIO'08, Eindhoven, The Netherlands, 2008, WeD3.
16. L. Cai, Y. Kang, H. Hu, *Optics Express*, **24**, 4640 (2016).
17. D. Tulli, D. Janner, M. Garcia-Granda, R. Ricken, V. Pruneri, *Appl. Phys. B*, **103**, 399 (2011)
18. S. Tonchev, M. Kuneva, *Proc. SPIE*, **3950**, 246 (2000); BG Patent, No. 63788, Bull. No.12, 29.12.2002.

ТЕХНОЛОГИЯТА ПРОТОНЕН ОБМЕН ЗА ОПТИЧНИ ВЪЛНОВОДИ В LiNbO₃ И LiTaO₃ – КРАТЪК ОБЗОР

М. Кънева

Институт по физика на твърдото тяло „Акад. Г. Наджаков”, бул. Цариградско шосе 72, 1784 София

Постъпила на 10 октомври 2016 г.; коригирана на 20 ноември, 2016 г.

(Резюме)

Литиевият ниобат (ЛН) и литиевият танталат (ЛТ) са нелинейни, двойно лъчепрекупващи кристали, които са от важно значение за оптоелектрониката. Най-забележителното в тях, освен отличните оптични качества, са високите им електрооптични коефициенти ($r_{33} > 30.5 \text{ pm/V}$), които позволяват светлинното поле да бъде лесно управлявано с помощта на електрически сигнали. Протонният обмен е метод за получаване на слоеве от LiM_{1-x}N_xO₃ (M = Nb, Ta) върху подложка от LiMO₃. Обменените слоеве имат съществено по-висок показател на пречупване (n_e) за необикновения лъч отколкото непротониран кристал (промяната в показателя на пречупване след протониране е $\Delta n_e = 0.120-0.150$ за ЛН и $\Delta n_e = 0.015-0.020$ за ЛТ при дължина на вълната $0.633 \mu\text{m}$). Поради това те проявяват силно вълноводно и поляризиращо действие. Целта на настоящата публикация е обобщение на основните технологични модификации на метода и приложението му за различни модулатори, използвани като основна част на съвременните оптоелектронни устройства (навигационни и комуникационни системи, биодатчици и др.)