

Optical and Structural Properties of Proton Exchanged MgO:LiNbO₃ waveguides

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We show that proton exchanged MgO:LiNbO₃ optical waveguides exhibit complex structural chemistry which is different from known for congruent material. It's shown that Annealing Proton Exchange process leads to strong degradation of surface region of waveguides, but Soft Proton Exchange preserves the crystal structure of MgO:LiNbO₃.

Keywords: proton exchange, magnesium doped lithium niobate, waveguide, crystal structure.

Introduction

Lithium niobate, LiNbO₃, is very important ferroelectric crystal with various acoustical, integrated optical and optoelectronic applications. Integrated optical devices require material improvements in optical homogeneity and optical damage resistance. It is known [1-3], that resistance to optical damage in LiNbO₃ is effectively improved if more than 4.5 mol.% MgO is added to the melt.

One of basic methods to make optical waveguides in LiNbO₃ crystals is proton exchange (PE) [4]. Despite the simplicity of the proton exchange technique, the PE LiNbO₃ waveguides exhibit very complex structural chemistry. Our works [5,6] have allowed us to identify seven different crystallographic phases, β_4 , β_3 , β_2 , β_1 , κ_2 , κ_1 and α -H_xLi_{1-x}NbO₃, which can be realized in the PE LiNbO₃ waveguides. Annealing is an important parameter in the fabrication of PE waveguides. Actually, most of the published results were obtained using PE waveguides, whose index profiles indicate that they are in the α -phase. These waveguides can be fabricated by two step process, including proton exchange and subsequent annealing (APE waveguides [7]) or by one step process, so-called Soft Proton Exchange (SPE) [8,9]. In contrast to APE LiNbO₃, the SPE LiNbO₃ waveguides preserve nonlinear coefficients of bulk material [10].

Several fundamental issues in the fabrication of waveguides by proton exchange in MgO:LiNbO₃ have not been addressed to date. Previously published investigations of PE MgO:LiNbO₃ waveguides have been incomplete and restricted to particular fabrication process [11-15].

Up to date, there are no reports about crystallographic study on proton exchange layers in Mg:LiNbO₃. In the present paper we shall show that structural and optical properties of the different types of PE MgO:LiNbO₃ waveguides depend strongly on fabrication conditions.

Preparation of samples and their characterization

PE waveguides were fabricated in optical grade X-cut MgO:LiNbO₃ substrates, containing 5.5 mol.% of MgO, supplied by Crystal Technology, Inc.

As the source of PE we used benzoic acid whose acidity can be reduced by adding up to 5 wt.% of lithium benzoate. Postexchange annealing at 330°C was used to modify the parameters of the waveguides. HTPE process [9] has been performed in the mixture of stearic acid and lithium stearate. Soft Proton Exchange process has been realized by HTPE in melts with lithium stearate concentration higher than the threshold value. In this case uniform α -phase waveguides are formed.

In order to find modes effective indices, we measured excitation angles of dark m-lines of modes with one prism coupler setup at the 633 nm wavelength. The refractive index profiles throughout the guide depth were reconstructed by the IWKB technique [16]. The strains perpendicular to the substrate surface, ε''_{33} , were calculated from rocking curves on plane (220) obtained by double-crystal X-ray diffractometer DRON-3 (Cu K α_1 radiation, Si (311) monochromator):

$$\varepsilon''_{33} = -\Delta\theta_{hkl} \cdot ctg\theta_{hkl} \quad (1)$$

where θ_{hkl} is the Bragg angle of the reflected surface plane for stress-free substrate.

Experimental results

Fig.1 shows the surface extraordinary index increase, Δn_e as a function of strain ε''_{33} for X-cut MgO:LiNbO₃ substrates. One can see that this structural phase diagram is absolutely different from such diagram, obtained for congruent material [6]. No doubt that some MgO:H_xLi_{1-x}NbO₃ phases exist, however, in contrast to structural phase diagram for congruent material [6], the possible phase transitions for PE MgO:LiNbO₃ are not so evident.

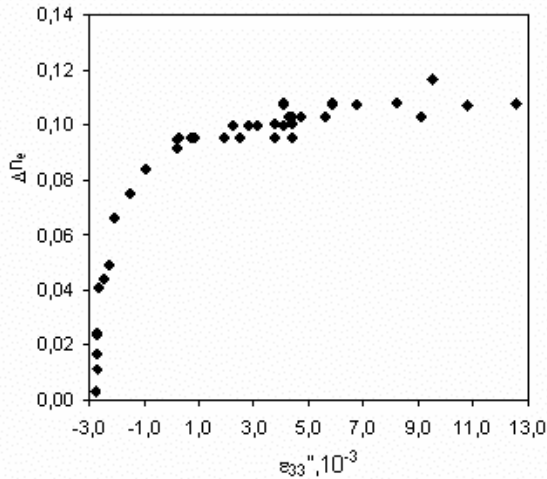


Fig.1. Dependence of the extraordinary index variation Δn_e measured at $\lambda=633$ nm versus the surface value of deformation ε''_{33} normal to surface plane.

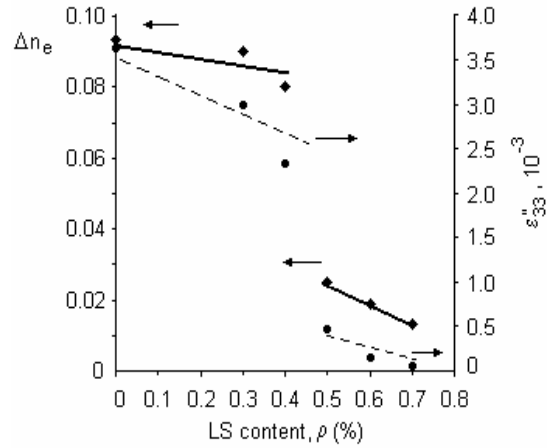


Fig.2 Refractive index change (rectangular dots, solid curve) and strains ε''_{33} (circular dots, dotted curve) as a function of melt composition in stearic acid + lithium stearate melts at 370°C.

Fig.2 shows the variation of the MgO:H_xLi_{1-x}NbO₃ surface value refractive index increment, Δn_e , and deformations ε''_{33} as a function of the LS content for HTPE at 370°C. The two regions can be clearly recognized. These regions correspond to, at least, two different phases. The Δn_e and ε''_{33} are found to be largely independent of melt composition for dilution up to threshold value, ρ_0 (Fig.2). The α -phase MgO:H_xLi_{1-x}NbO₃ fabricated at $\rho > \rho_0$ is characterized by low index increment and deformations.

The rocking curves on plane (220) and refractive index profiles for X-cut PE MgO:LiNbO₃ samples obtained under different exchange and annealing conditions are presented in Fig. 3.

As exchanged waveguides, similar to PE waveguides in congruent LiNbO₃, are characterized by positive deformations ε''_{33} . Three peaks can be clear recognized at X-ray rocking curve (Fig.3a). Most probably, these three peaks correspond to three different phases in PE MgO:LiNbO₃. However, after some time of annealing, ε''_{33} becomes negative. It is possible to obtain waveguide with large index increment ($\Delta n_e = 0.1$) and with zero deformation ε''_{33} (Fig.3b). Future annealing leads to increasing of absolute value of negative deformation.

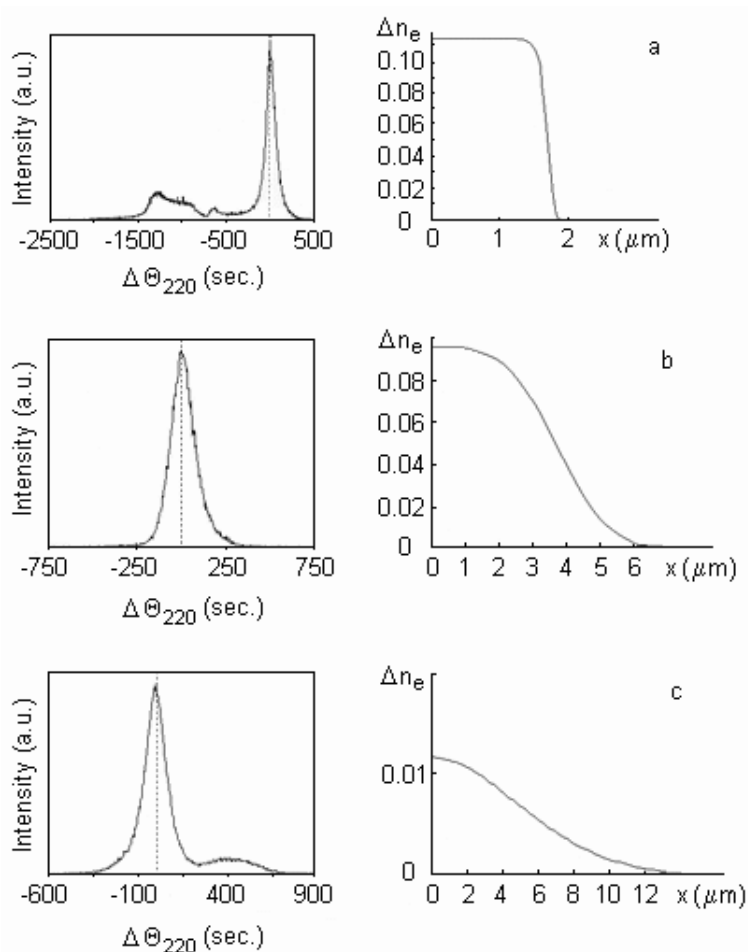


Fig.3. Evolution of X-ray rocking curves and refractive index profiles for PE MgO:LiNbO₃ sample, fabricated at benzoic acid melt at 220°C for 6 h with annealing at 330°C.

a) as-exchanged sample; b) after 17.5 h of annealing; c) after 462 h of annealing.

fabricated by APE technology (Fig.3c), the α -phase MgO:H_xLi_{1-x}NbO₃ prepared by HTPE technology characterized by absence of distorted surface layer. Therefore, the HTPE process preserves the crystal structure of α -phase MgO:LiNbO₃ waveguides. It is expected that such waveguides would show lower optical losses than APE waveguides.

Conclusion

The crystal structure and optical properties of PE MgO:LiNbO₃ structures are strongly depended on proton exchange and annealing conditions. Some crystal phases MgO:H_xLi_{1-x}NbO₃ can be generated on surface of MgO:LiNbO₃ single crystals depending on exchange and annealing conditions. It has been shown that traditional for congruent LiNbO₃ crystals APE technology leads to formation of distorted layer at the surface of MgO:LiNbO₃. It is expected that it would drastically increase the optical losses in APE MgO:LiNbO₃ waveguides. In contrast, the HTPE MgO:LiNbO₃ waveguides do not contain any distorted layer at the surface region. Therefore, such waveguides should show lower propagation losses.

It is very surprising that well annealed APE Mg:LiNbO₃ waveguides contain distorted layer with deformations $\epsilon''_{33} \approx -3 \cdot 10^{-3}$. Rocking curves for APE samples with $\Delta n_e < 0.1$ show broad additional peak (Fig.3c). Precise polishing shows that thickness of this layer is very close to thickness of initial as-exchanged layer.

It was obtained that this distorted layer is not been removed even after long reverse proton exchange process in LiNO₃-KNO₃-NaNO₃ melt. IR spectral measurements show that after RPE process at 330°C during 15 h, the intensity of -OH absorption at 3485 cm⁻¹ becomes similar to one for initial MgO:LiNbO₃ crystal, however the rocking curve shows additional peak (Fig. 4) with even larger deformation than that for initial APE sample.

Therefore, one can conclude that APE process leads to structural damage of surface layer, that can be a reason of additional scattering of light in APE MgO:LiNbO₃ waveguides.

Fig.5 shows the X-ray rocking curve for HTPE MgO:LiNbO₃ sample fabricated at 370°C for 5 h at the melt with $\rho=0.7\%$. One can see, that in contrast to α -phase MgO:H_xLi_{1-x}NbO₃

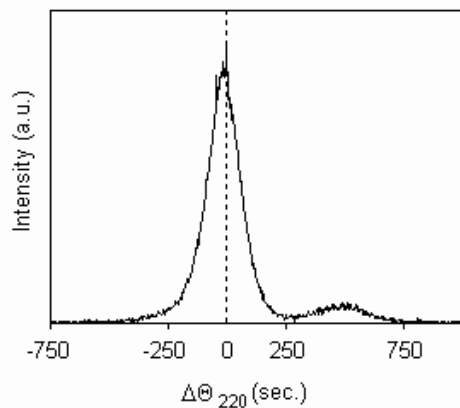


Fig.4 X-ray rocking curve for PE MgO:LiNbO₃ sample, fabricated at benzoic acid melt at 220°C for 6 h then annealed at 330°C for 552 h and treated at the melt LiNO₃-KNO₃-NaNO₃ at 320°C for 24 h.

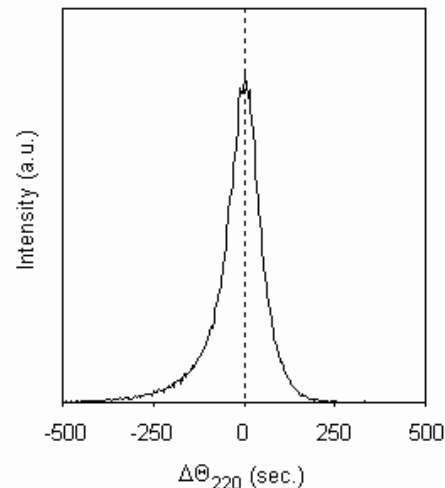


Fig. 5. X-ray rocking curve for HTPe MgO:LiNbO₃ sample fabricated at 370°C for 5 h at the melt with $\rho=0.7\%$.

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