Spontaneous polarization reversal induced in α-phase lithium niobate channel waveguides by proton exchange

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The α-phase waveguides are well known for preserving both the excellent nonlinear properties and the ferroelectric domains orientation of lithium niobate substrates. However, by using the piezoresponse force microscopy (PFM), we present a coherent study on ferroelectric dipoles switching induced by the one step fabrication process of α-phase waveguides on Z-cut congruent lithium niobate (CLN) substrates.

Keywords: *α-phase lithium niobate waveguides; piezoresponse force microscopy; spontaneous polarization reversal; piezoelectric coefficient*

INTRODUCTION

It is already known that waveguides fabricated on periodically poled lithium niobate (PPLN) substrates are one of the most widely used devices for many nonlinear optical applications based on the quasi-phase matching (QPM) process. However, an efficient nonlinear process such as second harmonic generation (SHG), spontaneous parametric down conversion (SPDC) or different variants of optical frequency conversion, requires, among others, waveguide fabrication techniques that allow preserving both the nonlinear coefficient and the domains orientation of the substrate. Despite the large number of articles that have proven the efficiency of α-phase channel waveguides for optical frequency conversion in PPLN devices, it has been observed that even α-phase waveguides in PPLN can exhibit uncontrollable nanodomains switching, which can be harmful to the initial periodic structure and obstruct the (QPM) process. Having in mind this idea, we performed, a coherent study of ferroelectric dipoles switching that occurred during the fabrication of α-phase channel waveguides on Z-cut congruent lithium niobate (CLN) substrates, by using the piezoresponse force microscopy (PFM).

The α-phase waveguides were fabricated by using High Vacuum Proton Exchange (HiVacPE) technique [1]. The proton exchange process was performed in a hermetically sealed hourglass tube. A mixture composed of Benzoic Acid (BA) and Lithium Benzoate (LB) was used as proton source. The waveguides were fabricated using different LB concentrations ranging from 2.5% up to 3% incremented by 0.1%, at T=300°C for t=72 hours. In this concentration range, the major difference between the HiVacPE and Soft Proton Exchange (SPE) is the very high reproducibility and control of the index contrast obtained by HiVacPE technique [2]. As expected, the α-phase waveguides exhibit an exponentially decreasing index profile and low values of the index contrast [1]. These kind of profiles and the calculated values of index contrast are the signature of lower $H^+ \leftrightarrow Li^+$ substitution ratio (<10%) [3, 4].

RESULTS

The PFM investigations were carried out on the xy plane by respecting the geometry of the samples as depicted in Fig 1. The topography and piezoresponse of the sample surface were simultaneously probed, the PFM microscope software providing three overlaid images namely, topography (exactly like a classical atomic force microscope),

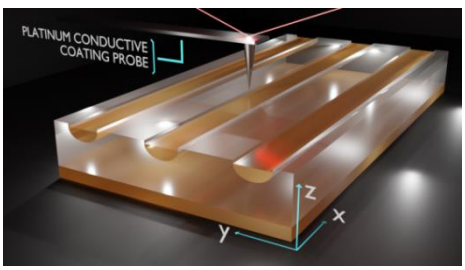

Fig. 1. Experimental setup for PFM investigations of α-phase channel waveguides. Cantilever movements are in x-y plane with the respect of samples geometry allowing scanning both waveguides and virgin CLN regions in a single scan.

PFM magnitude, and PFM phase, respectively, all of them associated to the same investigated area. Therefore, Fig. 2 shows 2D images recorded on α-phase channel waveguides fabricated with ρ_{LB}=2.5% in the bath. The images

correspond to: (a) topography, (b) PFM magnitude, (c) PFM phase, and (d) is the corresponding line profiles extracted at the same location of the images triplet.

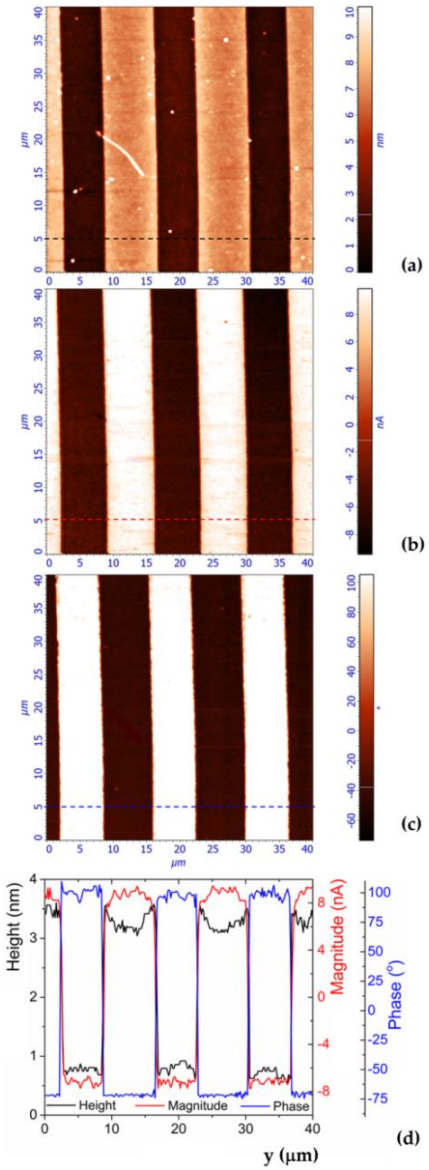

Fig. 2. Images triplet (topography, PFM magnitude and PFM phase) of CLN sample protonated with 2.5% LB: (a) topography of the sample, (b) 2D of PFM magnitude mapped in the same location as topography, (c) PFM phase of the piezoresponse mapped in the same location as topography and (d) line profiles extracted at the same location of the images triplet.

For quantitative assessments of the piezoelectric-like response of protonated regions (waveguides), we carried out the surface displacement calibration procedure of the piezoresponse signal [5]. Since this procedure depends on the quality of the calibration of the z-scanner itself, we have used a test PPLN sample with a known piezoelectric coefficient. Doing so, we were able to identify the conversion factor (γ_{AFM}) linking the amplitude of the vibration of the surface and the PFM response of the surface, respectively. Therefore, from the amplitude value h of the piezoresponse signal measured in a given point of the sample surface and knowing the amplitude V_{DC} of the probing *DC* voltage, the local piezoelectric coefficient d_{33} was determined from the slope of the plot of Eq. (1) [5].

$$h = \gamma_{AFM}\, d_{33} V_{DC} \tag{1}$$

The reduction in the piezoelectric coefficient d_{33} induced by the proton exchange compared with the virgin CLN is ranged between 18.3% for 2.5% LB in the bath and 8.8% for 3% in the bath. We found a very good agreement between index contrast (optical investigation) and d_{33} reduction (PFM investigations) being proof of a clear connection between the increasing of the refractive index (Δn_e) and the decreasing of the magnitude of spontaneous polarization (ΔP_s) in protonated regions.

Having a high interest towards nonlinear performances of photonics devices based on PPLN substrates, we have also investigated how deep the spontaneous polarization reversal induced by proton exchange takes place inside the α-phase channel waveguides. Therefore, in order to evaluate this depth, all the samples were subjected to a

clever surface engineering. The surface containing the channel waveguides was polished at a small angle ϑ with the respect of the xy surface plane. The angle, measured under optical microscope setup is less than ϑ=0.10°±0.05°. This allows the observation by PFM of the evolution of spontaneous polarization reversal as the waveguide thickness decreases from the surface down into the substrate. Fig. 3 provides a sketch of the experimental configuration and geometrical details. After scanning of a 50×50µm^2 area, the sample has moved in x-axis direction so that the next scan imagines the same waveguides but with decreasing thickness as the scan progresses.

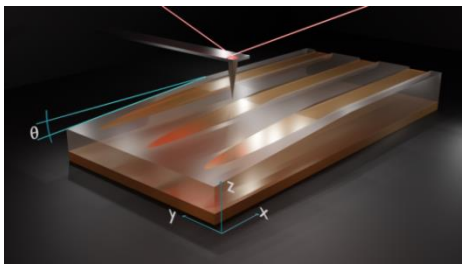

Fig. 3. α-phase waveguides polished with the respect to the xy surface for revealing the deepness of polarization reversal.

Our investigations reveal that the whole deepness of spontaneous polarization reversal induced by proton exchange is 5±0.2 nm. All investigated samples exhibit, in the limit of measurements errors, the same deepness of spontaneous polarization reversal.

DISCUSSION

By corroborating PFM investigation and optical characterizations of α-phase protonated regions and virgin CLN on ±Z surfaces of the samples, we find a very good agreement between index contrast (optical investigation) and d_{33} reduction (PFM investigations). We clearly showed that the increase in the in-diffused proton concentration (increase in index contrast) in protonated zones decreases the piezoelectric coefficient d_{33} values. We estimated a decrease between 18.5% and 9% depending on the acidity of the bath. This fact was experimentally demonstrated only for high $Li^+ \leftrightarrow H^+$ substitution ratio (>70%) in the so-called β-phase waveguides [6]. To the best of our knowledge, this is the first time that such a study has been conducted on α-phase waveguides for which the substitution ratio is less 10%. Also, we have assessed that the spontaneous polarization reversal takes place only in a 5±0.2 nm very thin subsurface layer. This result is important for any devices using α-phase channel waveguides in PPLN. In such a case, the 5±0.2 nm subsurface layer of the waveguide can no longer assure the QPM process. In this case, two scenarios can appear. First, if the propagation of different optical modes (associated to pump, signal, and idler, respectively) takes place mainly towards the surface of the waveguides, which is the case for waveguides exhibiting very low index contrast $<1.5\times10^{-2}$, the QPM is disturbed and the devices will exhibit poor performance of the SHG process. Second, if the propagation of different optical modes takes place mainly in the middle of the waveguides, which is the case for waveguides exhibiting index contrast $>1.5\times10^{-2}$, the QPM is not disturbed and SHG or SPDC processes can proceed successfully.

Therefore, beyond basic research aspects, our work contributes to the applied research effort on frequencies conversion in waveguides fabricated on LN platforms, where generation and manipulation of photons are crucial since real-world implementations demand high efficiency devices.

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