Fabrication of SPE waveguides on PPLN: formation of nanodomains and their impact on the SHG efficiency

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In this paper, we present the material characterization that has been made to explain the poor nonlinear performance observed in certain channel waveguides produced by soft proton exchange in periodically poled congruent lithium niobate (PPLN) crystals. The study was performed using complementary methods of domain visualization: piezoelectric force microscopy and confocal Raman microscopy. It has been shown that the waveguide fabrication process can induce the formation of the structure of needle like nanodomains, which can be responsible for the degradation of the nonlinear response of the waveguides created in PPLN crystals.

Keywords: soft proton exchange, periodical poling, nanodomain structure, second harmonic generation

Short title: Nanodomains in SPE waveguides formed on PPLN

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1. Introduction

Lithium niobate (LN) single crystals with tailored periodical domain structure are widely used for second harmonic generation (SHG) and other nonlinear optical applications based on realization of the quasi-phase matching (QPM) effect [1-3]. The historically first periodically poled crystals with waveguides for SHG were created on congruent LN [4-6]. In order to fabricate the efficient sources of photon pairs in the telecommunication windows, the parametric down-conversion process in waveguides caused by periodicity of the nonlinear coefficient to fulfil QPM conditions was used [7]. The waveguides were fabricated using the soft proton exchange (SPE) process in periodically poled lithium niobate (PPLN) single crystals [8]. The index profile reported in Fig. 1 was measured using the m-lines technique and a He-Ne laser in planar waveguide formed on the polar surface [9].

The SPE waveguides were used for fabrication of the highly efficient components for quantum signal processing and quantum communication [10, 11]. Nevertheless, according to our experience, the nonlinear performance of these devices sometimes is much lower than expected values. For example, the sample being studied in the present paper has the record value of the optical loss (ranged from 0.15 to 0.5 dB/cm) for this type of devices, but demonstrates disappointing nonlinear performance. In the SHG experiments the obtained conversion efficiency maximum is as low as 10 % W⁻¹ cm⁻², which is significantly less than 140 % W⁻¹ cm⁻² already demonstrated in SPE waveguides [9]. Moreover, the shapes of SHG spectra for the most of the waveguides (Fig. 2) are quite different from the theoretical sinc². In this paper, we present and discuss the results of the experimental studies we have undertaken to explain the mentioned above unexpected behavior and to reveal the technological parameter, which has to be controlled to avoid these poor performances.

The study performed by means of complementary experimental methods Piezoresponse Force Microscopy (PFM) and Confocal Raman Microscopy (CRM) allowed us to reveal the appearance of a number of the surface nanodomains during the waveguide fabrication. The nanodomain structure superimposed with the periodic bulk domains existing in PPLN wafers could be responsible for the observed degradation of the nonlinear performances.

2. Experimental

The samples under investigation were prepared on a 0.5-mm-thick Z-cut congruent LN wafer, which was periodically poled using the E-field technique with liquid electrodes and a photoresist mask with periods ranging between 15.6 and 16.6 μ m [6]. The SPE procedure was carried out in benzoic acid bath with 2.9 % lithium benzoate at 300 °C for 3 days, using a SiO₂ mask to fabricate the channel waveguides along the X axes. The waveguide with width ranging from 4 to 8 μ m was fabricated on the side of the sample representing Z⁺ polar surface before periodical poling. The planar waveguide used for index profile determination was produced on the opposite Z⁻ polar surface.

PFM was used for obtaining the high-resolution images of the static domain patterns on the wafer surface [12] (Fig. 3).

3. Results and discussion

In Fig. 3a, it can be seen that SPE waveguide fabrication process does not modify noticeably the piezoelectric properties of the poled area (Z^- orientation), while in the originally oriented areas (Z^+ orientation) the clear contrast of the waveguide was obtained by PFM. Higher magnification allowed us to reveal that the boundary between waveguide and untreated area represented a stripe filled by submicron nanodomain dots (Fig. 3b). Similar patterns were observed for the surface nanodomain structures, which appeared in front of moving domain wall in LN crystals with the surface layer modified by proton exchange [13-15]. Going to the higher resolution, it is possible to visualize them as in the waveguide so in

the dense structure of isolated nanodomains with lateral sizes down to 50 nm (Fig. 4).

It is known that isolated nanodomains in CLN possess the needle-like shape with aspect ratio typically ranging from 50 to 100 [16] (see Fig. 5). Therefore, it is reasonable to assume that the nanodomains extend into the crystal bulk down to the depth ranged from 2.5 to 5 μ m. This means that in the Z⁺ area, the whole waveguide volume is affected by these nanodomains. It looks like the nanodomain structure in the Z⁻ area is absent (Fig. 5).

CRM allowed us to visualize the domain structure in the crystal bulk [17, 18]. Fig. 6 presents the cross-section along the waveguide, which crosses the stripe domain with two domain walls. It is seen clearly that the domain walls reach the crystal surface. This fact confirms that the periodical domain structure, which is necessary to fulfil QPM conditions, is preserved. It was shown experimentally, that the similar surface nanodomain structures appeared under the action of the pyroelectric field during fast heat treatment of LN crystals [19, 20].

Further investigations are in progress to confirm that the observed nanodomain structures are produced by a heat treatment, and that the degradation of the SHG spectra and the reduction of the conversion efficiency are caused by existence of the surface nanodomains in the Z^+ area of the PPLN crystal. The additional study will be done for understanding the role of nanodomain structure appeared in the surface layer of the waveguide area. According to our previous studies [21], we do not expect a modification of the Raman spectrum due to the presence of SPE waveguide.

4. Conclusion

We present the material characterization that has been made to explain the poor nonlinear performance observed in certain channel waveguides produced by soft proton exchange in periodically poled congruent lithium niobate crystals. The study performed by means of piezoresponse force microscopy and confocal Raman microscopy allowed us to reveal the appearance of a number of the surface nanodomains during the waveguide fabrication. These nanodomains are superimposed onto periodic bulk domains of PPLN wafers; thus, they can be responsible for the degradation of the nonlinear performance.

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Figure captions

Fig. 1. Extraordinary index profile for $\lambda = 633$ nm of the studied waveguide fabricated by the SPE technique.

Fig. 2. Typical SHG spectrum obtained in SPE waveguides with low conversion efficiency.

Fig. 3. PFM images of the PPLN domain structure (along Y direction) crossed by SPE waveguides.

Fig. 4. Nanodomains at the border and in SPE waveguide produced on the Z^+ surface.

Fig. 5. (a) PFM view of the surface at the interface between Z^+ and Z^- area; (b) a scheme of the interface cut showing the shape of the surface nanodomains appeared at Z^+ area.

Fig. 6. Image of Y-cut face of PPLN obtained by CRM demonstrating that the domain walls in PPLN still reach the surface after waveguide fabrication.

Fig. 1.





Fig. 2.



Fig. 3.



Fig. 4.



Fig. 5.



Fig. 6.