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SHORT COMMUNICATION

GROWN PERIODICALLY POLED LITHIUM NIOBATE CRYSTAL: PERIOD STABILIZATION

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High degree of the period stabilization and homogeneity of the regular domain structure of the Nd:Mg:LiNbO$_3$ crystals was obtained in the growth process by the Czochralski method from melt with lithium dioxide excess, in the direction along the normal to the $\{01\bar{1}2\}$ close packed face.

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Bulk periodically poled LiNbO$_3$ (PPLN) single crystal is a promising material for nonlinear optical devices using quasi-phase-matching (QPM) [1]. Active periodically poled lithium niobate crystals are prospective for producing compact multifunctional laser devices because of combination of the advantages (merits) of QPM-generation harmonics [2-4], optical parametric oscillation [5], the capability of realization of self-doubling frequency [6, 7] and more complicated processes, for example, the summing of the frequencies.

Uniform micron-scale periodic structures with flat domain boundaries are needed for QPM devices. Doping with neodymium was used earlier to form periodically poled domain structure (optical superlattice) on the basis of rotation-induced growth striations in the bulk LiNbO$_3$ single crystal [8, 9, 10]. The goal of this work is to increase the stability of the domain structure period (several microns dimension) of the PPLN crystal with flat domain walls. Previously, it was reported that stable periodicity on the 3 mm crystal length was obtained [11].

Bulk periodically poled domain structure with flat domain walls was created in Nd:Mg:LiNbO$_3$ crystal in the growth process by the Czochralski method in the direction along the normal to the close packed $\{01\bar{1}2\}$ face [4, 8, 9, 12]. The Fig. 1 shows the positions of the face relative to the crystallographic X, Y, Z-axes.

The introduction of impurities during the crystal growth has a periodic character under asymmetric heat field and $\kappa_{\text{eff Nd}} < 1$ (“rotating“ growth striation). The ferroelectric domain boundaries are connected with the extrema of impurity modulation [12].

The face $\{01\bar{1}2\}$ on the growth front was about 10 mm for boules 20 mm in diameter and, therefore, in the central part of boule domain walls coincided with the face. Nevertheless, generally, in these crystals the domain period variations were observed. These variations are connected with the change of the heat removal in growth chamber in the process of the crystal pulling out. In addition, smooth increasing of period was observed as crystal diameter was increased due to melt level lowering. As a consequence, the period stability was ~ 2% for 1-3 mm crystal length.

Periodically poled Nd:Mg:LiNbO$_3$ single crystals with (4.4±0.1) μm period and flat domain walls (Fig. 2) were grown on the Czochralski installation along the normal to the $\{01\bar{1}2\}$ face , i.e.

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along $<0\bar{5}52>$ direction. The seed was cut from the slice of YZ-cut at an angle of 57^{o} with respect to the Z-axis (Fig. a, b). Starting melt contained lithium dioxide surplus in relation to congruent compound (2:1, about 60 mol % Li_2O, see diagram of the Li_2O- Nb_2O_5 system [13]) and, consequently, the crystal growth was obtained by flux method. Before, Li_2O excess was used for the growth of facets at $LiNbO_3$ crystals [5], and K_2O was added for growth of the homogeneous single domain $LiNbO_3$ crystals [6]. The period of domain structure was determined by the rate ratio crystal pulling/rotation. The crystal pulling velocity was 5 mm/h, the rotation velocity was 22 rpm (correspondingly, calculated period was 3.8 μm), and platinum crucible diameter was 50 mm.

The ferroelectric domain structure (Fig. 1) on the polished X-cut was studied by selective chemical etching [12]. After etching, the inspection of sample surface was made using a metalographic microscope. The concentration of Nd and Mg impurity was measured using wave dispersive X-ray microanalysis.

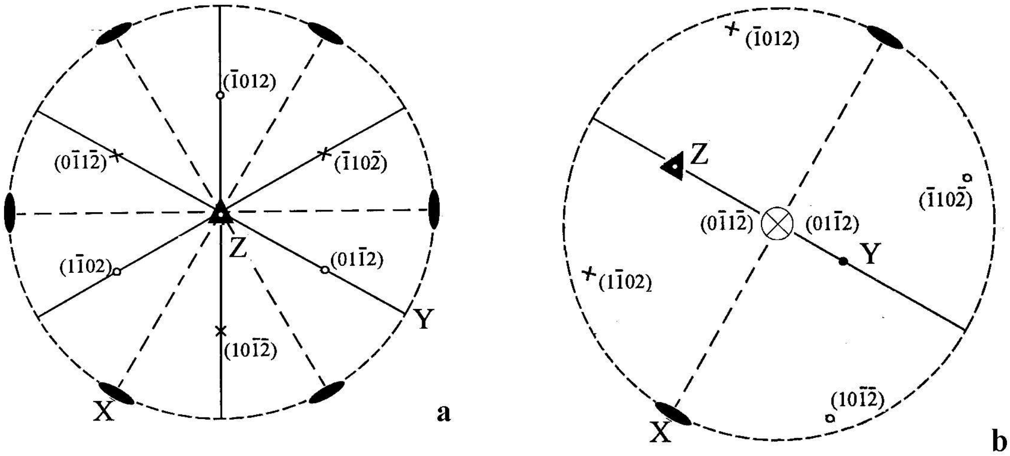

Fig. 1. The stereographic projection of the lithium niobate crystal point group ($\bar{3}m$) and $\{01\bar{1}2\}$ face projections; X, Y, Z-crystallographic axes; a: the projection center is Z-axis; b: the projection center-growth direction is $<0\bar{5}52>$.

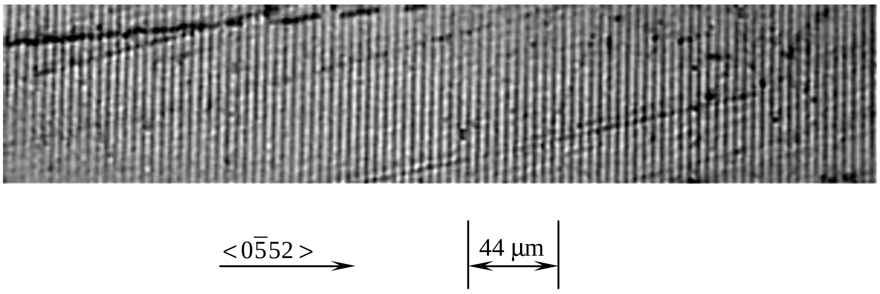

Fig. 2. Periodic domain structure on the X-cut of Nd:Mg:$LiNbO_3$ crystal, revealed by selective chemical etching.

In the growth process we reduced the heater power (Fig. 3, curve 1). The crystal diameter and face dimension gradually increased as far as crystal was pulled out from melt (Fig. 3, curves 2, 3). Nevertheless the domain structure period remained enough constant (Fig. 3, curve 4). Average relative measuring error was 2% on the 20 mm length. The measuring of the all magnitudes was implemented from the crystal bottom. It is remarkable that the domain structure of the crystals obtained by flux method is free from typical defects, as e.g. single domain regions, usually having the dimensions about several ten microns. The impurity concentration in the grown crystal was determined by X-ray micro-analysis method: Nd_2O_3- 0.5 wt %, MgO- 0.8 wt %.

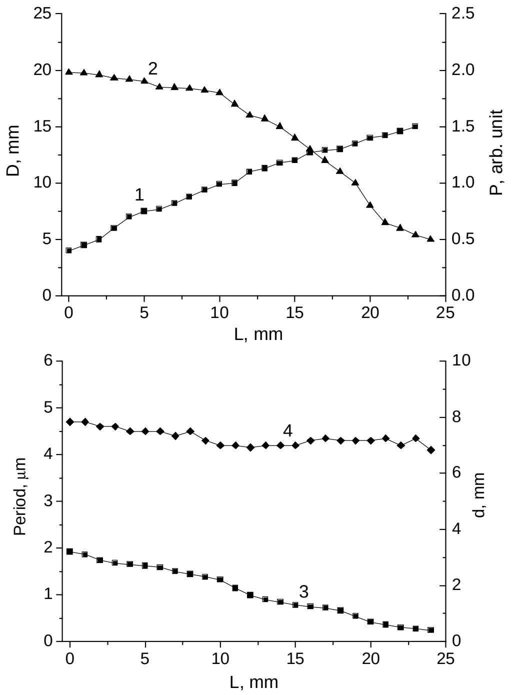

Fig. 3. The dependences of the growth process of Nd:Mg:LiNbO$_3$ crystal on length: the indication of the heater power scale (curve 1); crystal diameter (curve 2); face dimensions (curve 3); the period of regular domain structure (curve 4).

Earlier, we have shown that the ferroelectric domain walls coincide with the maxima and minima of Nd-impurity modulation in the PPLN grown along the normal to the face [12]. The “effect of the face” reveals itself in the increase of the neodymium incorporation in the crystal and in the creation of smooth domain boundaries [12]. The $\{01\bar{1}2\}$ face cuts the convex growth isotherm and grows by means of consecutive precipitation of the layers by the advanced steps (the tangential growth mechanism).

In the growth process from flux (at Li$_2$O excess) the impurity (Nd^{3+}) introduction mechanism changes and becomes self-regulated. We observed high stability of the regular domain structure: (4.4±0.1) µm on 20 mm of the crystal length in the growth direction. On the one hand, flux method suggests the increasing (in comparison with growth from melt) of the thickness boundary layer on the growth front, which becomes less sensitive to the variations of the temperature conditions. On the other hand, there are possible changes of the profile and impurity modulation depth.

We suppose that high stability of the regular domain structure is connected with the impurity (Nd^{3+}) introduction mechanism in the growth process from flux (Li$_2$O excess) and the presence of the close packed $\{01\bar{1}2\}$ face on the growth front.

Periodically poled active nonlinear Nd:Mg:LiNbO$_3$ crystal with domain structure period 4 µm and 7 mm length along the normal to the $\{01\bar{1}2\}$ face was used in the experiment on the intracavity QPM self-frequency summing:

$$\omega_{\text{laser}} + \omega_{\text{pump}} = \omega_{\text{sum}}$$

In this process diode laser (λ = 0.81 µm) was the pump of active medium, whereas unabsorbed part of the diode laser radiation was involved in the process of nonlinear interaction with laser generation wave (λ = 1.084 µm). At the output of the crystal the radiation wave of the summing frequency (λ=0.464 µm) was registered [14]. The flat domain walls make possible wide aperture interactions of laser beams in bulk PPLN crystals.

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