Formation of Stripe Domain Structures by Pulse Laser Irradiation of LiNbO₃ Crystals

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The formation of quasi-regular stripe nanodomain structure induced by ultraviolet pulse laser irradiation was studied in single crystalline congruent lithium niobate LiNbO₃. It has been shown that the domain structure with average period about 4 µm consists of “straight” and “wave-like” nanodomain rays. The scanning electron microscopy allows us to reveal that the rays represent quasi-periodic arrays of individual nanodomains with average size 50–200 nm and period 100–500 nm. The spatial frequency multiplication of quasi-regular structures has been revealed.

Keywords Lithium niobate; nanoscale domain structures; pulse laser irradiation; laser heating; pyroelectric field

I. Introduction

Formation of periodically poled domain patterns in single crystalline lithium niobate LiNbO₃ (LN) has been studied actively due to numerous applications such as quasi-phase-matched (QPM) nonlinear-optical devices, electro-optic Bragg deflectors, photonic bandgap structures, and piezoelectric devices [1]. Most of commonly used techniques of electric-field-induced domain inversion present problems when periods of a few microns and below required for first-order QPM non-linear processes at blue and near-ultraviolet wavelengths are needed. In order to overcome the limitations imposed by electric-field poling, the technique of light-assisted electric-field poling (LAP) which takes advantage of the ultraviolet (UV) light-induced essential decrease of the coercive field has been developed during the past few years for lithium tantalate [2, 3] and lithium niobate [4–6] crystals.

The surface domain inversion method has been developed recently [7]. In this method the interaction of the intense UV laser light with single-domain LN leads to fabrication of shallow self assembled domain structure consisting of quasi-periodic stripe domains with sub-micron width and few micron periods [8–16]. It is clear that for practical application of the proposed method it is necessary to study the peculiarities of the domain formation as a result of pulse UV laser irradiation. The mechanisms responsible for polarization reversal process under such unusual conditions are to be specified. This is the reason which makes
us to undertake detail investigation of the processes of UV-induced formation of stripe domain structure in LN.

II. Experiment

The polarization reversal procedure involved the pulse laser illumination of the Z+ polar face of the congruent LN (CLN). The samples under investigation represented 1 mm thick optically-polished CLN single domain wafers cut normal to polar axis.

It should be noted that existence of the needle-like residual domains in the wafer can essentially affect the laser-induced domain growth. So, it was absolutely necessary to be sure that the initial domain state of the studied samples in our experiments was single domain. For this purpose, the studied CLN wafers were subjected to careful examination before the irradiation procedure. An optical visualization of residual domains was performed by polarization microscopy without application of electric field. It has been shown that the choice of optimal microscope aperture allows us to visualize the domains existing both at the surface and in the bulk of the wafer in transmitted as well as in reflected modes. All used wafers were checked to be free of optically visualized initial domains.

The frequency-quadrupled Nd:YAG laser (Brilliant, Quantel, France) operating at wavelength 266 nm with up to 50 mJ pulses of 4 ns duration was used as the source of pulse laser radiation. The measurements of the pulse laser energy have been made by average power and laser radiation energy meter.

The obtained nanodomain structures have been revealed by very shallow chemical etching in pure HF during 5–10 minutes at room temperature. The obtained surface relief corresponding to existing domain structure was visualized by optical microscopy and scanning electron microscopy (SEM). Statistical analysis of the obtained images has been carried out.

III. Results and Discussion

It has been shown experimentally that high-intensity pulse laser irradiation of the surface of CLN single crystals leads to formation of complex nanodomain structures [7]. In our recent articles, we have proposed the qualitative explanation of the origin of the switching field produced by laser pulse [9–15]. According to our model, the irradiation by laser pulse leads to rapid heating of the sample surface with corresponding rapid decrease of spontaneous polarization. The retardation of screening of the depolarization field change leads to appearance of the pyroelectric field which suppresses the polarization reversal (“suppression field”). The cooling of the heated volume, which starts after the ending of the laser pulse, leads to increase of the spontaneous polarization. The pyroelectric field appeared during cooling due to screening retardation of increasing depolarization field induces the polarization reversal (“switching field”).

Systematical analysis of the light-induced domain images allows us to distinguish three spatially separated types of the domain structures at Z+ polar surface: 1) isolated nanodomains, 2) quasi-regular stripe domain structures, and 3) self-similar domain structures [9–14].

Isolated nanodomains were generated mostly along the boundary of the irradiated zone. Such behavior can be explained assuming higher cooling rate in this region as compared with the zone center. According to the suggested model, the magnitude of pyroelectric field is proportional to the cooling rate. In this case, the magnitude of the induced local pyroelectric field can be high enough for arising of the new domains (nucleation).
In the central part of the irradiated zone the quasi-regular and self-similar domain structures were observed. It has been revealed using optical and scanning probe microscopy (SPM) that these structures consist of narrow domain stripes (domain rays). The average ray width strongly depends on the irradiation conditions and varies from 150 to 400 nm [10]. As the cooling rate within irradiated zone is considered to be essentially lower than at the zone boundary, the switching pyroelectric field in this region can initiate only growth of already existing domains.

As periodically poled LN is used in various nonlinear-optical devices we would consider UV-induced quasi-regular structures in detail. Analysis of the surface domains reveals that the quasi-regular stripe domain structures started to form at the boundary of the irradiated zone and grew to the zone center. Three groups of stripe domain structures were observed. The stripes in the given group were oriented predominately along one of three Y crystallographic directions (Fig. 1a).

The average period of the formed quasi-regular stripe structures was about $3.9 \pm 0.8 \mu m$ [9]. The experiments did not reveal any dependence of the structure period on the density of the laser pulse energy.

In order to verify our assumption that the growth of quasi-regular structure started from already arisen individual domains formed at the irradiated zone boundary, the following experiment was carried out. The polar surface with artificial micro-scale scratch has been subjected to intensive UV irradiation. It is known that the scratches induce arising of the nucleation centers. Experiment has shown that the Y oriented domain ray growth starts from the scratch (Fig. 1b). The averaged domain structure period in this case was $3.9 \pm 1.8 \mu m$.

It has been shown by us while studying the domain structure formation in wide range of experimental conditions that the domain growth strictly along Y directions is obtained only in those irradiated regions where induced pyroelectric fields were relatively low [14]. In our experiments, practically parallel straight strictly oriented domain rays were observed in the central part of irradiated zone. Nevertheless, some rays demonstrate wave-like behavior (Fig. 1). The detail study of the domain images allows us to assume that the straight and wave-like domain rays appear at the different time intervals after pulse laser irradiation (different stages of domain pattern formation).

The obtained experimental results were attributed to the essential spatial inhomogeneity of pyroelectric field appearing during cooling after the UV induced heating of the crystal surface and its variation with time. The time dependence of the pyroelectric field during heating/cooling cycle induced by pulse laser irradiation has been simulated according to

![Figure 1. Quasi-regular stripe domain structures formed in LN by UV pulse laser irradiation: a) perfect surface quality; b) artificial micro-scale scratch inside the illuminated zone.](image-url)
our proposed model taking into account the retardation of bulk screening of depolarization field. The results of theoretical estimations of temporal behavior of pyroelectric field in the center of irradiated zone during cooling after the ending of the laser pulse are presented in Fig. 2. According to calculated temporal behavior of the pyroelectric field, we can propose the following scenario of the domain structure formation consisting of three stages.

During the first stage, the appearance of the individual domains along the edge of the irradiated zone or along the micro-scale scratch occurred. The calculations support this assumption predicting higher values of the pyroelectric field in these areas.

During the second stage, the appeared individual domains gave rise to the growth of the straight domain to the center of the irradiated zone. This growth occurred in lower pyroelectric fields due to difference in cooling rates between zone center and its boundary. In this case the field value is enough only for realization of the domain growth strictly along Y direction assuming that along this direction the magnitude of nucleation threshold field is minimal.

The third stage starts when the increasing field overcomes the threshold for domain growth in directions other than Y. Such conditions lead to strong deviations from the straight line growth and to realization of the wave-like domain growth.

The low concentration of the individual domains along the edges leads to growth of straight domains in low field and formation of the quasi-periodic structure of “first generation” with comparatively large period. The subsequent cooling leads to further increase of the pyroelectric field at the zone boundary. Thus, the concentration of the individual domains increases and stimulates the growth of “second generation” stripe domains in the space between the existing ones. This process decreases essentially the structure period. At the same time, the shape of new growing domains changes from straight to wave-like in the cause of time. This effect can be attributed to the increase with time of pyroelectric field in the central zone due to cooling of the whole sample. So, at some moment the given domain of second generation in high enough field breaks off the straight movement.

**Figure 2.** Simulated qualitative time dependence of the pyroelectric field after pulse laser irradiation switch off (cooling cycle) in the center of irradiated zone (arbitrary unites).
It has been shown by statistical analysis that the average distance between neighboring straight domains of first generation is $7.1 \pm 3.9 \, \mu m$ (Fig. 3a), while the average period of the whole quasi-regular domain structures including the domains of second generation is about $3.9 \pm 1.8 \, \mu m$ (Fig. 3b).

The distribution of period of quasi-regular domain structure was fitted by Gaussian function. It is important to point out that the normalized half-width of Gaussian function of $(w/\Lambda)$ decreased from 0.55 to 0.46 after appearance of the domains of the second generation, where w is a half-width of Gaussian function, $\Lambda$—average period of domain structure.

The high resolution visualization technique realized by SEM allows us to demonstrate that the individual ray represents array of isolated nanodomains (Fig. 4). Statistical analysis of the domain images shows that sizes of the individual nanodomains range from 50 to 200 nm with distances between neighboring nanodomains ranged from 100 to 500 nm.

It has been determined that straight nanodomain arrays are formed as a result of consecutive arising of new nanodomain strictly along $Y$ crystallographic direction. In contrast, the wave-like domains arrays are formed as a result of regular deflection of arising nanodomains from $Y$ direction.

Multiple pulse laser irradiations allow us to realize the anisotropic growth of isolated nanodomains mostly along $Y$ direction with subsequent merging and formation of continuous domain rays.

**Figure 3.** Distribution functions of periods (a) for domains of “first generation”, (b) for all domains in quasi-regular domain structures.

**Figure 4.** SEM images of (a) “straight” and (b) “wave-like” nanodomain arrays in CLN single crystals.
IV. Conclusions

We have studied in details the quasi-regular stripe domain structure generated in CLN single crystals by high-intensity UV pulse laser irradiation. The considered model of polarization reversal under the action of pyroelectric field allows us to explain the main experimental results. The proposed time dependence of the pyroelectric field has been verified by computer simulation. The effect of spatial frequency multiplication of quasi-regular structures due to coexistence of two different types of stripe domains (“straight” and “wave-like”) has been revealed. This effect was attributed to increasing of pyroelectric field during cooling, which led to arising of the wave-like stripe domains of the “second” generation. It has been shown by SEM that stripe domain structure represents arrays of isolated nanodomains with average size 50–200 nm and period 100–500 nm.

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