Self-organized nanodomain structures arising in lithium tantalate and lithium niobate after pulse heating by infrared laser

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Formation of nanodomain structures during cooling after pulse heating by infrared laser has been studied in congruent lithium tantalate (CLT) and lithium niobate (CLN) single crystals. *In situ* study of the domain structure evolution allowed to reveal that in CLN the isolated domains appeared at the edges of the irradiated zone and grew to the center. In contrast, the quasi-regular stripe domain structure appeared in CLT near the edge of irradiated zone. The difference has been attributed to lower Curie temperature and larger pyroelectric coefficient of CLT. The self-organized domain structures can be used for periodical poling with submicron periods.

Keywords: domain engineering, nanodomains, pyroelectric field, quasi-regular structures, pulse laser heating

Short title: Domain structures after laser heating in CLN & CLT

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1. Introduction

The modern development of telecommunication technologies requires fabrication of submicron- and nano-scale regular domain structures (domain engineering) for creation of improved quality optical components, such as Bragg reflectors, beam steering devices, and narrow band filters [1-3]. Backward second harmonic generation, when the wave with the fundamental frequency propagates in opposite direction to its second harmonic one [4, 5], needs periodically poled crystals with submicron period gratings with nanoscale accuracy of period reproducibility.

Uniaxial ferroelectrics lithium niobate $LiNbO_3$ (LN) and lithium tantalate $LiTaO_3$ (LT) are the most popular materials for domain engineering [6]. The periodically poled LN and LT (PPLN and PPLT) are used for laser light frequency conversion based on quasi-phase matching effect [7-10]. They look like the best candidates for nanodomain engineering [11]. The fabrication of the tailored domain structures is realized usually by application of electric field and needs photolithography for electrode patterning [12]. This method is inefficient for precise domain structuring with period below 2 μm [13-15].

The formation of the stable quasi-regular nanodomain structures in congruent LN (CLN) and congruent LT (CLT) crystals under highly non-equilibrium switching conditions was demonstrated experimentally [16-19]. The irradiation of the polar surface of Z-cut CLN plate by pulsed ultraviolet laser leads to formation of the shallow domain structure with the domain depth about few microns [20, 21]. In contrast, the irradiation by pulsed infrared (IR) laser initiates the formation of the bulk domain structure consisting of oriented nanodomain rays with depth up to 200 μm under the action of pyroelectric field [22]. It has been proved experimentally that the polarization reversal occurs during cooling after heating termination [23]. The increasing of plate temperature before laser irradiation leads to decrease of the domain density and to increase of growth anisotropy [24]. The simulation of the time dependence of the pyroelectric field during heating-cooling cycle taking into account the temperature dependence of the bulk conductivity and threshold field allowed us to reveal the temperature hysteresis and to explain all the observed effects [23, 25].

Similar effects have been obtained in CLT after pulse laser irradiation. The CLT specific feature is the appearance of three-dimensional maze-type structure in the central part of the irradiated zone heated above Curie temperature (T_C = 610 ^{o}C) [26].

In this work, we report the experimental study of the formation of self-organized nanodomain structures in CLT and CLN after pulse heating by IR laser for various pulse durations. The time dependence of the temperature measured during heating-cooling cycle was used for explanation of the domain structure evolution.

2. Experimental

The studied samples represented optically polished 0.5-mm-thick Z-cut plates of CLT (Oxide Corp., Japan) and 1-mm-thick CLN (SIPAT Co., China). The pulsed CO_2 laser with a wavelength 10.6 μm and Gaussian distribution of laser energy density was used for the sample heating by single pulse with duration ranged from 0.3 to 8 ms and energy ranged from 20 to 370 mJ. The laser irradiation was focused on Z+ polar surface by ZnSe lens with focal distance 51 mm. The distance between lens and plate surface was 14 mm.

For visualization of the domain structure evolution simultaneously with recording of the irradiated zone temperature the sample surface was covered by stripe periodic metal applications (80-nm-thick Cr) with period 5 μm and width 1.5 μm. The fast *in situ* recording of the domain evolution has been achieved from the nonexposed side by CCD camera (FastVision LLC) with time resolution 2 ms (Fig. 1). The temperature recording in the central part of the irradiated zone (diameter 0.8 mm) was carried out by high speed infrared pyrometer Klaiber KGA 740-LO with time resolution 6 μs in spectral range 1.5-2 μm, which allowed measuring temperature in the range from 200 to 1000 ^{o}C. As LN practically does not emit in the needed spectral range, the temperature recording can be achieved by measuring of radiation from metal stripes covering the polar surface.

The static stable domain structure appeared after irradiation was revealed by chemical etching using pure HF. The typical etching time for CLN is about 5 min and for CLT – 30 min [27]. The surface relief corresponding to domain pattern was visualized by optical microscopy (Olympus BX51) and with high spatial resolution by atomic force microscopy (AFM, MFP-3D SA, Asylum Research). Statistical analysis of the domain images has been used for extracting the averaged parameters characterizing the domain pattern geometry: anisotropy of domain ray growth and period of the domain structure.

3. Domain structures in CLN formed after pulse laser heating

It was shown by us earlier that after pulse IR laser heating of CLN, the irradiated zone was covered by the net of the separated nanodomain chains and domain rays of micron-scale width named as "hatching effect" (Fig. 2) [23]. The similar domain patterns were observed after fast heating by current along the electrode deposited on the polar surface of MgO doped CLN [25].

We have studied the dependence of the domain pattern parameters on the pulse duration $\boldsymbol{\tau}$ of IR laser at fixed power density. It has been shown that increasing of pulse duration leads to increase of the radius R of the zone occupied by domain structure (Fig. 3a) and increase of the average distance d between domain rays (Fig. 3b). It is necessary to point out that the total length of domain rays *L* is almost independent on the pulse duration and equals to 25 mm.

The obtained dependences have been fitted by power low R,d($\boldsymbol{\tau}$) = A $\boldsymbol{\tau}^{b}$ (Fig. 3). The exponent of power for radius (b_R = 0.64) was twice larger than the one for averaged distance

($b_d = 0.32$).

Characterization of anisotropy of domain rays growth was carried out by statistical analysis of the optical images of domain structure (Fig. 2). Analyzed zone has been divided into concentric ring-shaped regions with equal width. The distribution function of rays' orientation has been calculated for each region by approximation of the domain pattern by broken lines and calculation of the distribution function using SIAMS Photolab software. The fraction of Y-oriented domain rays (L_Y/L) was used for characterization of ray growth anisotropy. It has been shown that L_Y/L ratio remains constant in the central part and decreases near the edge of the irradiated zone (Fig. 4).

4. Domain structures in CLT formed after pulse laser heating

It was shown by us earlier that pulse laser irradiation of CLT led to formation of two zones with qualitatively different types of domain structure (Fig. 5a): three-dimensional maze-type structure appeared in the central part of the irradiated zone heated above Curie temperature (T_c = 610 ^{o}C) and self-similar structure consisting of domain chains formed in the vicinity of the edge of the irradiated zone [26]. The narrow ring-shaped transition zone of isolated nanodomains appeared between them.

We have studied the dependence of the domain pattern parameters on the pulse duration of IR laser at the fixed power density. It has been shown that the zone with self-similar structure appears at pulse duration about 1 ms and expands with its increasing (Fig. 6a). The zone with maze-type structure appears at pulse duration about 2 ms. The further increasing of pulse duration leads to increase of the radii of both zones (Fig. 6a). At the same time, the area of ring-shaped zone with self-similar structure remains constant (Fig. 6b).

Increasing of power density by decreasing of focusing distance leads to essential increasing of the growth anisotropy of nanodomain chains at the edges of irradiated zone.

The statistical analysis of the AFM domain images obtained for low and high power density was made for quantitative characterization of the self-similar structures (Fig. 5b). The angular segment of self-similar structure zone has been divided into stripe regions of equal width. For each stripe region, the histogram of the chain orientation has been calculated (Fig. 7). The anisotropy of chain growth direction in given region has been characterized by inverse value of the dispersion, which has been obtained as a result of fitting by Gaussian function.

For low power density 22 W/mm^2, the anisotropy of the chain growth direction in zone with self-similar structure is almost independent on the distance from the center of irradiated zone (Fig. 8a). In contrast, the anisotropy of the chain growth direction is increased for high power density 155 W/mm^2 from interior to external edge of the zone with self-similar structure (Fig. 8b). The domain chains near the interior edge of the zone were oriented with equal probability along three Y crystallographic directions with low anisotropy (Fig. 7a). Approaching to the external edge of the zone leads to increasing of the growth anisotropy and prevalence of the chain orientation along radius (Fig. 7b). The region with chain orientation along radius only and highest anisotropy was obtained near the external edge of the zone (Fig. 7c), thus leading to formation of strictly oriented quasi-regular domain structure [6].

5. Evolution of the domain structure in CLN during cooling after pulse laser heating

The time dependence of the surface temperature in the center of irradiated zone during and after pulse laser heating was measured for CLN and CLT plates covered by Cr applications. It was shown that the temperature of CLN and CLT changed almost equally for any pulse duration. Increasing of the pulse duration leads to increasing of maximum temperature, which exceeds the Curie temperature of CLT (about 610 ^{o}C) for low power density (22 W/mm^2) at pulse duration above 8 ms (Fig. 9). In this case the heating and cooling rates can achieve 10^5 ^{o}C/s.

The used technique allowed us to visualize the domain structure evolution in CLN only. The instantaneous images of domain pattern taken at different moments are presented in Fig. 10. The corresponding moments are marked on the measured time dependence of temperature in the center of irradiated zone (Fig. 11).

It is seen that the domain structure evolution starts during cooling after termination of the laser pulse. For 8 ms pulse duration polarization reversal starts after 13 ms following the termination of heating at the temperature about 330 ^{o}C, when the cooling rate is about 2×10^{4} ^{o}C/s (Fig. 11). The domain formation process starts with the appearance of isolated domains and subsequent growth of domain rays along periodical Cr electrodes oriented along given Y direction. The isolated domains arise at the distance 500 μm from the center of irradiated zone and the domain rays grow to the center. The total time of domain structure formation is about 90 ms. The switching process terminates, when the temperature of the central part of the sample is below 200 ^{o}C.

6. Discussion

The formation of self-organized nanodomain structures in CLN and CLT can be attributed to the action of the pyroelectric field E_{pyr}, which appears during cooling of the surface layer after termination of the IR laser pulse heating [23, 28]. The polarization reversal can occur only at the cooling stage at temperatures, when the E_{pyr} value overcame the value of the temperature dependent threshold field E_{th}. The analysis of the domain structures appeared in LN in wide field range shows that the ray growth anisotropy decreases with increasing of E_{pyr} [29]. According to this, we can conclude that in case of irradiation of CLN and CLT by IR pulse laser with low power density 22 W/mm^2 the spatial distribution of E_{pyr} is almost uniform over the whole irradiated zone. It has been demonstrated for CLT that increasing of the power density and decreasing of pulse duration leads to nonuniform distribution of E_{pyr} with maximum in the center of irradiated zone and to formation of quasi-regular strictly oriented domain structure at the edge of irradiated zone. This effect is due to higher cooling rate in the center of irradiated zone.

The *in situ* temperature recording has shown that both heating and cooling rates are equal for CLN and CLT. Thus, the qualitative difference of domain structures formed in these crystals under pulse laser irradiation in the same experimental conditions can be attributed to lower value of Curie temperature for CLT, which has been achieved during heating in the central part of the irradiated zone. Moreover, the larger pyroelectric coefficient of CLT leads to higher value of pyroelectric field.

7. Conclusion

The domain structure formation induced by pulse infrared laser heating in CLT and CLN single crystals has been investigated. The dependence of the domain structure parameters on laser pulse duration has been measured for both materials. The *in situ* study of the domain structure evolution in CLN with simultaneous temperature recording in the center of irradiated zone has been carried out during heating-cooling cycles. It has been shown that the polarization reversal starts during cooling after the pulse termination by appearance of the isolated domains at the edges of irradiated zone and their subsequent growth to the center. Characterization of the anisotropy of domain rays growth was carried out by statistical analysis of the optical images of domain structure. For high power density the anisotropy of the chain growth direction in CLT increased from interior to external edge of the zone with self-similar structure. The region with quasi-regular stripe domain structure oriented along radius was obtained near the external edge of the zone. The *in situ* temperature recording showed that the temperature of CLN and CLT changed almost equally during heating-cooling cycles under the same experimental conditions. The qualitative difference of domain structures formed in these crystals under the pulse laser irradiation can be attributed to lower value of Curie temperature and larger pyroelectric coefficient for CLT. The obtained formation of the stable domain structures under the action of the pyroelectric field can be used for domain engineering in the crystals of LN and LT family. This method can be especially useful for production of the PPLN and PPLT with submicron periods.

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Figure captions

Fig. 1. Experimental scheme of simultaneous in situ visualization of the domain structure evolution and recording of the irradiated zone temperature.

Fig. 2. Typical domain structure in CLN appeared after IR laser heating with pulse duration 2 ms and low power density 22 W/mm^2.

Fig. 3. Dependences on pulse duration of (a) radius of zone occupied by domain structure and (b) average distance between domain rays fitted by power law. Low power density: 22 W/mm^2.

Fig. 4. Dependence of the fraction of Y-oriented domain rays LY/L over the ring-shaped region on the distance from the center of domain pattern. Low power density: 22 W/mm^2.

Fig. 5. Typical domain structure in CLT appeared after laser heating with pulse duration 350 μs and high power density 155 W/mm^2: (a) optical and (b) AFM images. Domain pattern was revealed by chemical etching.

Fig. 6. Dependences of (a) radii and (b) areas of two zones (blue squares – zone with self-similar domain structure, red circles – zone with maze-type structure) on pulse duration. Low power density: 22 W/mm^2.

Fig. 7. Angle distributions of chain growth direction: (a) at the internal edge, (b) at the center, and (c) at the external edge of self-similar structure zone.

Fig. 8. Dependence of the anisotropy of chain growth in zone with self-similar structure on distance from the center of irradiated zone. Power density: (a) 22 W/mm^2, (b) 155 W/mm^2.

Fig. 9. (a) The measured time dependences of temperature in the center of irradiated zone for different pulse duration and (b) the dependence of maximum temperature on pulse duration for CLN plate. Power density: 22 W/mm^2.

Fig. 10. The instantaneous images of domain structure for different moments: a) 8 ms, b) 12 ms, c) 16 ms, d) 22 ms, e) 28 ms, and f) 34 ms, during cooling after pulse laser heating of CLN plate covered by Cr applications. Pulse duration 8 ms, power density 22 W/mm^2.

Fig. 11. Measured time dependence of the temperature in the center of irradiated zone. Red lines mark moments corresponding to the instantaneous images of domain structure in Fig. 10.