Ultrashort-pulse optically-assisted domain engineering in lithium niobate

C. E. Valdivia, C. L. Sones, S. Mailis, J. D. Mills, R. W. Eason

Shortened Title:

fs optically-assisted poling in lithium niobate

Abstract:

Ultrashort laser pulses (~150-200 fs) of near-ultraviolet (305 nm) to near-infrared (800 nm) wavelengths have been used to optically-assist domain nucleation and growth in lithium niobate. Within illuminated areas, the electric field required for domain nucleation is reduced by up to 41% in undoped and up to 98% in 5-mol% Mg-doped congruently melting materials, allowing direct-writing of inverted domains with electric fields as small as 100 V mm^{-1}. A first step towards the formation of optically-defined periodically poled lithium niobate was achieved by illumination via a phase mask, demonstrated over small areas with a period of 5.25 microns.

Precise engineering of the ferroelectric domain structure of materials such as lithium niobate (LN) is required for applications such as quasi-phase-matched (QPM) nonlinear optical interactions [1]. This is even more essential for advanced functionality such as near-ultraviolet generation, backward propagating harmonic generation, or electro-optically controlled Bragg gratings where the periods required span the micron to sub-micron scale. Conventional domain engineering techniques, typically electric-field poling, permit the fabrication of gratings with periods as small as several microns [2]. This involves photolithographically defining a periodic electrode on one face of the ferroelectric material and applying a large electric field exceeding the coercive field, E_c, of the material. Therefore, this technique requires a new mask for defining structures of each specific period and cleanroom facilities for the lithographic process, adding to the cost and complexity of fabricating such a device. Therefore, alternative methods of forming such devices with greater flexibility and less expense are currently under investigation.

All-optical poling is one such alternative domain inversion technique which, if possible, would remove the need for photolithography and simplify the fabrication process. In this process, ultraviolet pulses of nanosecond-duration were focused onto the +z face of LN with fluences above the ablation threshold, but in the absence of an applied electric field [3]. To date, surface domains of 150-300 nm widths have been formed, but truly periodic structures have been technically challenging to produce.

An intermediate technique between conventional electric-field poling and all-optical poling is *light-assisted poling* (LAP), the process of simultaneously applying a uniform electric field and patterned light to define and induce a bulk or surface domain-inverted structure within a ferroelectric crystal. Such a technique removes the need for photolithography and may push the boundaries of minimum period length. Furthermore, the

use of interference can eliminate the need for new masks whenever a new periodic pattern is required. Fujimura *et al.* [4] demonstrated that the simultaneous application of 1.75 kV and ultraviolet light from a mercury lamp patterned by an amplitude mask could be used to define domain-inverted structures in magnesium-doped lithium niobate (Mg:LN) with a period $\Lambda = 15.4$ μm. This was further characterized by Wengler *et al.* [5,6] using 305- and 334-nm cw light, obtaining reductions in the effective coercive field in regions of illumination reaching 50% in LN doped with MgO in concentrations greater than the optical damage threshold. Visible light from a cw Ar^+ laser has been tightly focused using a confocal microscope to directly-write surface domain patterns in LN lightly doped with Er^{3+} ions by Dierolf and Sandmann [7]. Finally, Sones *et al.* [8] showed that cw visible light can produce up to a 31% reduction in the electric field for nucleation of domains in undoped congruently melting lithium niobate (CLN), and up to an order of magnitude reduction in CLN doped with MgO above the optical damage threshold.

The LAP investigations discussed above focused solely on cw light, using visible and ultraviolet wavelengths. The intention of this paper, therefore, is to present our results using a femtosecond laser system producing ultrashort pulses of duration ~150-200 fs. An optical parametric amplifier allowed the tuning of wavelengths from ultraviolet (UV) to near-infrared (NIR), including 305, 334, 364, 383, 400, 514.5, and 800 nm. These wavelengths were chosen to compare with cw LAP experiments conducted in the UV [6] and visible [8] ranges. Following the results of these previous experiments, two types of crystals have been investigated. The first is undoped congruently melting lithium niobate (CLN). The second is 5-mol% MgO-doped congruently melting lithium niobate (Mg:CLN), with a doping level above the optical damage threshold which is desirable due to its improved resistance to photorefractive damage [9].

The setup used for these light-assisted poling experiments is shown in Figure 1 and is described in greater detail in Ref [8]. In brief, the z-cut LN sample, oriented for illumination on the $-z$ face, is held between transparent silica windows by two silicone O-rings. Water electrodes are confined by the O-rings on the surface of the samples, allowing the application of high electric fields for domain inversion with and without illumination. Low-intensity polarized red light from a HeNe laser passed through the crystal along a path collinear with the femtosecond beam. This red light was then imaged onto a CCD camera, revealing domain inversion through stress-induced birefringence at the domain walls via a crossed polarizer, and electro-optic refractive index contrast when an electric field is present. Prior to all LAP measurements, conventional "dark" electric-field poling was used to cycle the crystals through forward and reverse domain inversion to determine the coercive field, E_c (electric field required for macroscopic domain inversion and growth), and the dark nucleation field, $E_{I=0}^{nucl}$ (electric field required for the nucleation of domains in the absence of light), for each sample. This process also avoids the first poling cycle, which is well-known to have a higher coercive field than any subsequent cycle.

During illumination, domain inversion was monitored optically throughout the poling process and also subsequently, by chemical etching in hydrofluoric acid (HF). All wavelengths attempted, from UV to NIR, were able to form domain-inverted structures over a range of intensities with an electric field below the dark nucleation field, forming domains within the illuminated areas only. Figure 2 shows the normalized nucleation field as a function of average intensity for wavelengths, $\lambda = 305$, 334, and 514.5 nm, incident on Mg:CLN which has a dark nucleation field $E_{I=0}^{nucl} \sim 6.3$ kV mm^{-1}. Here, the femtosecond laser system provided a repetition rate of 1 kHz and 250 kHz for UV and visible wavelengths, respectively. Illumination of Mg:CLN by wavelengths $\lambda = 334$ and 514.5 nm shows a remarkable decrease of up to 98% of the electric field required to nucleate domains using

conventional electric-field poling in the absence of illumination, permitting domain formation at voltages as low as 50 V (E = 100 V mm^{-1}). In undoped CLN, with a dark nucleation field $E_{I=0}^{nucl} \sim 21$ kV mm^{-1}, the effect of illumination is lessened but still achieves a 41% reduction in the nucleation field using λ = 400 nm light.

Light-assisted poling can be used for direct-writing of inverted domain structures. Biasing 0.3-mm thick CLN crystal at 15000 V mm^{-1}, a beam of wavelength λ = 400 nm was scanned across the sample to form patterns with arbitrary shapes, not constrained to the hexagonal symmetry of the crystal. One such example, spelling the letters "ORC" with bulk domains extending throughout the thickness of the sample, can be seen in Figure 3. The light-induced domain lines grew quickly and uniformly without spreading beyond the illuminated region. Reaching 100-200 μm widths, these light-assisted features form bulk domains extending from the $-z$ face to the $+z$ face of the crystal [Figure 3 (a) and (b), respectively]. For these exposure conditions, the edges of the domain structure appear very jagged. In fact, investigation of the edges of the domain lines under the high magnification of a scanning electron microscope (SEM) shows that domain walls form straight edges along *arbitrary* directions and even *curved* lines (inset of Figure 3). Recent SFM measurements of inverted domains with circular shapes on the $-z$ face, formed by conventional electric-field poling, suggest that bulk domains will adopt crystalline symmetries along the three principle y-axes within a depth of approximately 30 μm or less, and as a result appear hexagonal upon reaching the $+z$ face [10]. However, despite the nucleation of these light-assisted domains on the $-z$ face and growth towards the $+z$ face, they do not necessarily appear hexagonal even on the $+z$ face, as shown in Figure 3 (b). This may represent a very useful ability of these light-assisted domains to break free of the crystalline symmetry confines and permit the formation of bulk domain structures with more ideally-shaped arbitrary patterns designed for specific application requirements.

While direct-write is a very flexible fabrication method, it is not appropriate for such structures as periodically poled lithium niobate (PPLN) where many thousands of precisely defined periodically domain-inverted regions may be required for high-conversion efficiencies of quasi-phase-matched nonlinear interactions. In this case, projection or interference techniques should be more suitable. To this end, a phase mask was introduced into the holder adjacent to the $-z$ face of the crystal, oriented with the grating k-vector parallel to the x-axis, as shown in Figure 1. Optimized for operation at $\lambda = 514.5$ nm, the phase mask produced an intensity grating with period $\Lambda = 5.25$ μm. A 0.5-mm thick Mg:CLN sample was then illuminated with this intensity grating while varying the exposure conditions of intensity, electric field, and illumination time, as shown in Figure 4. With much smaller dimensions than the ~100-μm sizes of directly-written examples discussed above, the illumination time became an extremely important parameter. For example, comparing Figure 4 (a), (b), and (d), the only exposure parameter that varied between these spots was illumination time, being 5 s, 3 s, and 1 s, respectively. In both Figure 4 (a) and (b), the light-assisted domains have spread laterally and merged together, forming a large central domain-inverted region. However, the exposure conditions in Figure 4 (d) appear to be at the point where adjacent domain lines are just beginning to merge. It should be noted that the illuminating beam has a Gaussian intensity profile, resulting in domains merging at the center and separating toward the edges. Together, the images of Figure 4 show a clear progression from over-exposure towards optimal conditions, where (f) has a clear separation of adjacent periods throughout nearly the entire beam width. The domain pattern, as revealed by HF etching, has precisely the same period as the illuminating intensity pattern, i.e. $\Lambda = 5.25$ μm.

In a hybrid direct-write/interference technique, scanning the beam across a static phase mask and sample can extend this fabrication method to a greater number of periods while maintaining the same intensity and electric-field parameters. Scanning speed would therefore

replace the illumination time as one of three important exposure parameters. Figure 5 shows various regions produced in Mg:CLN by scanning the beam at different speeds under exposure conditions of wavelength $\lambda = 514.5$ nm, intensity $I = 20$ W cm^{-2}, and electric field $E = 1200$ V mm^{-1}. Over-exposure causes merging of adjacent domains [Figure 5 (a)] and jagged edges as the domains spread outside the intensity peaks (b). However, under improved exposure conditions, high-quality periodic grating lines form [Figure 5 (c)], exhibiting straight domain walls aligned with the illumination pattern and having an inverted/non-inverted duty-cycle of ~25/75 (d). In the future it may be possible to also control this duty-cycle through precise control of all exposure conditions, obtaining an optimal 50/50 ratio. However, it should be noted that a 25/75 duty-cycle is already optimal for even-ordered QPM [11].

The selective choice of exposure conditions during LAP has resulted in the observation of some intuitive trends. The three important exposure parameters are intensity, electric field, and illumination time (or scanning speed, if appropriate), together defining the effective exposure. In general, the increase of any one of these parameters (or decrease of scan speed) individually will result in the further growth and/or spreading of a domain. However with a suitably small electric field, a light-assisted domain will grow to a maximum size as determined by the beam size, and thereafter appear to grow no further regardless of intensity or illumination time for the range of exposures tested. Under the lowest exposure conditions, only surface domains are formed but their depths are expected to be suitable for waveguide applications. Moreover, pushing surface domains deeper may be possible by increasing an appropriate exposure parameter. Even bulk domains extending from the $-z$ to $+z$ crystal faces do not necessarily follow the crystalline symmetry, as demonstrated by the directly-written structure of Figure 3, which may suggest an important advantage of LAP over conventional electric-field poling. In contrast, using high electric fields, light-assisted domains formed with

moderate to high intensities will tend to grow along the crystalline axes, and this transition point is typically near 1 kV for Mg:CLN of thickness 0.5 mm.

Shorter wavelengths appear to require lower intensities for similar reductions in the light-induced nucleation field. This can be broadly grouped into the UV, visible, and NIR regimes. Wavelengths in both UV and visible ranges have been shown to be capable of reducing the nucleation field to as small as 100 V mm^{-1} (

Figure 2) in Mg:CLN, but using UV light required approximately one-third the intensity of visible light. To date, the first experiments using light in the NIR range (λ = 800 nm) have produced up to an 81% reduction in the nucleation field of Mg:CLN, but used at least an order of magnitude higher power than required with visible light. Therefore, even though this light-induced process of reducing the nucleation field occurs over a broad wavelength range, there may be an optimum wavelength regime in terms of photon efficiency.

This ability to dynamically lower the nucleation field with light reduces the requirement for high voltage equipment. Recently there has been much progress in the development of very low coercive field stoichiometric crystals [12]. However, a permanently low coercive field limits the application of these materials in electro-optic devices because correspondingly low external electric fields can result in domain reversal. The advantage of LAP is that it can be applied to crystals with intrinsically high coercive fields, and hence any periodic or arbitrary domain structures will be stable upon the application of external electric fields below this much larger electric field.

Several comparisons can be made between the cw and ultrashort-pulse LAP processes. Firstly, the wavelength range demonstrated with ultrashort pulses now exceeds the range of all previous cw experiments, and includes the first demonstration of infrared light-assisted domain formation (λ = 800 nm). Secondly, ultrashort pulses permit the use of a further 5-fold reduction in the nucleation field as compared to cw illumination. Although the average

intensities used are of a similar order of magnitude to cw exposure, the peak intensities are 10 orders of magnitude higher. Thirdly, while cw light is more likely to form hexagonal structures that will spread laterally, ultrashort pulses can permit domain shapes and sizes as defined by the illumination pattern. Even bulk domains have been produced exhibiting jagged and rounded edges that do not conform to the crystalline symmetry on either –z or +z faces. Finally, cw LAP domains are typically visible via their stress-induced birefringence when using a crossed polarizer. However, ultrashort-pulse LAP often results in inverted domains not clearly visible with this technique, implying the generation of relatively little stress under low exposure conditions. In cw LAP experiments the domains suddenly appear visible when the nucleation field is reached, whereas in ultrashort-pulse LAP the domains often were not visualized optically until they were over-exposed. In these cases where optical visualization is not possible, selective HF etching must be relied upon to determine when domain inversion has occurred.

While ultrashort-pulse LAP presents a different operating regime than does cw LAP, it will be interesting to investigate any analogies to the difference between pulsed-voltage and conventional non-pulsed electric-field poling. In pulsed-voltage poling, short ms-duration high-voltage pulses first nucleate a domain, then subsequent pulses push the domain deeper towards the opposite face, as in Ref [13]. In pulsed LAP, similarly, it is possible that the short-duration high-intensity light pulses first nucleate a domain, then subsequent pulses push the domain deeper and spread it to fill the illumination pattern. In this case however, the voltage is constant, but the local nucleation field is reduced to below the applied electric field value, being modulated with each light pulse. Future experiments will investigate the effect of the number of pulses on the depth of light-assisted domains to determine if this analogy is appropriate.

The results presented here represent the first instance of ultrashort-pulse light-assisted electric-field poling. Illumination by a wide wavelength range of 305-800 nm has been demonstrated to form inverted domain structures with a voltage as small as 50 V, or electric field of 100 V mm^{-1}, in Mg:CLN. This effect has been used to form directly-written arbitrarily shaped surface and bulk domains, as well as forming periodic domain patterns by illumination via a phase mask. Future investigations will attempt to improve the quality of light-assisted poling and its applicability to useful devices. By converting the Gaussian beam into a top-hat profile, uniform exposure conditions will be attained across the entire spot and enable larger scanned areas. Phase masks with smaller periods may prove able to push towards smaller domain periods. In this case, measuring the depth of these small light-assisted surface domains will be essential in ensuring the optimal exposure conditions for optically periodically poled lithium niobate (OPPLN) used in future bulk and waveguiding applications.

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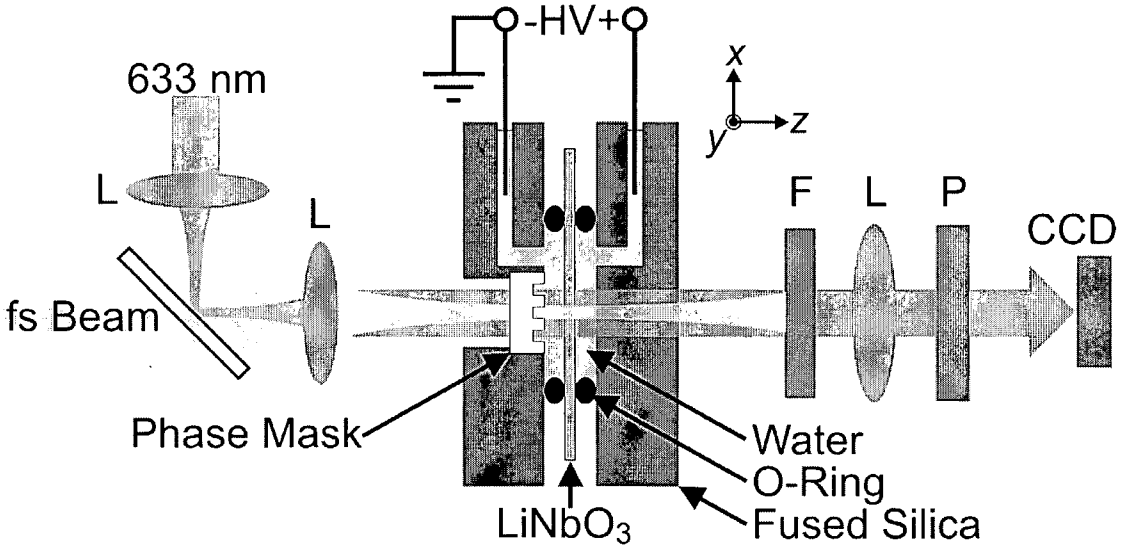

Figure 1: Light-assisted poling setup, used both with and without the phase mask. The coordinate system represents the orientation of the lithium niobate sample, showing the *k*-vector of the phase mask is parallel to the *x*-axis. The polarized beam of wavelength $\lambda = 633$ nm is of low intensity and used only for visualization of the poling process via the crossed polarizer and CCD camera. (F = red filter; L = lens; P = polarizer; HV = high voltage.)

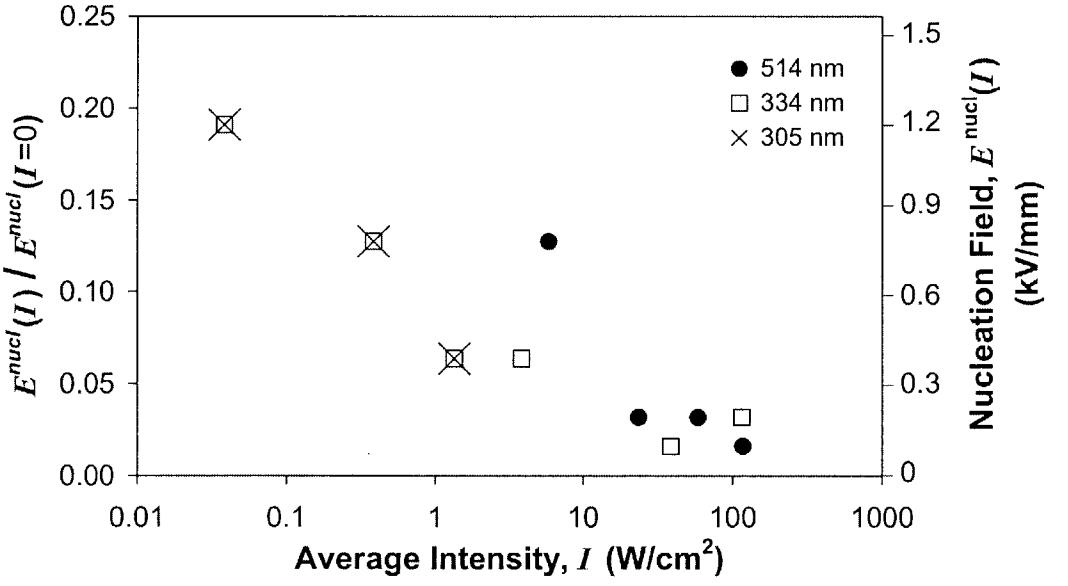

Figure 2: Intensity-dependent nucleation field [$E^{nucl}(I)$] normalized relative to the dark nucleation field [$E^{nucl}(I=0)$], plotted as a function of average intensity for ultraviolet (305, 334 nm) and visible (514.5 nm) wavelengths illuminating 5-mol% MgO-doped congruently melting lithium niobate. Larger intensities reduce the minimum electric field required for nucleation down to 100 V mm^{-1}. Further optimizations may push this to even lower electric fields for small surface domains.

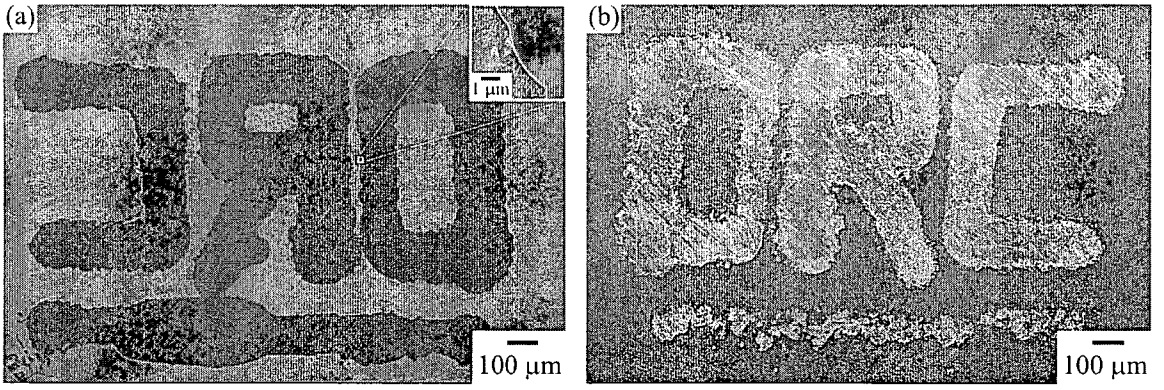

Figure 3: Direct-writing of domain structures in undoped congruently melting lithium niobate using $\lambda = 400$-nm light with an electric field of ~15000 V mm^{-1}, or 28% below the dark nucleation field. The light-patterned domains nucleated on –z face (a), but formed bulk domains by growing to the opposite +z face (b), shown here after 20 min HF etch. Domain walls follow the shape of the beam rather than the crystalline symmetry of the sample, exhibiting arbitrarily angled or circular edges (inset).

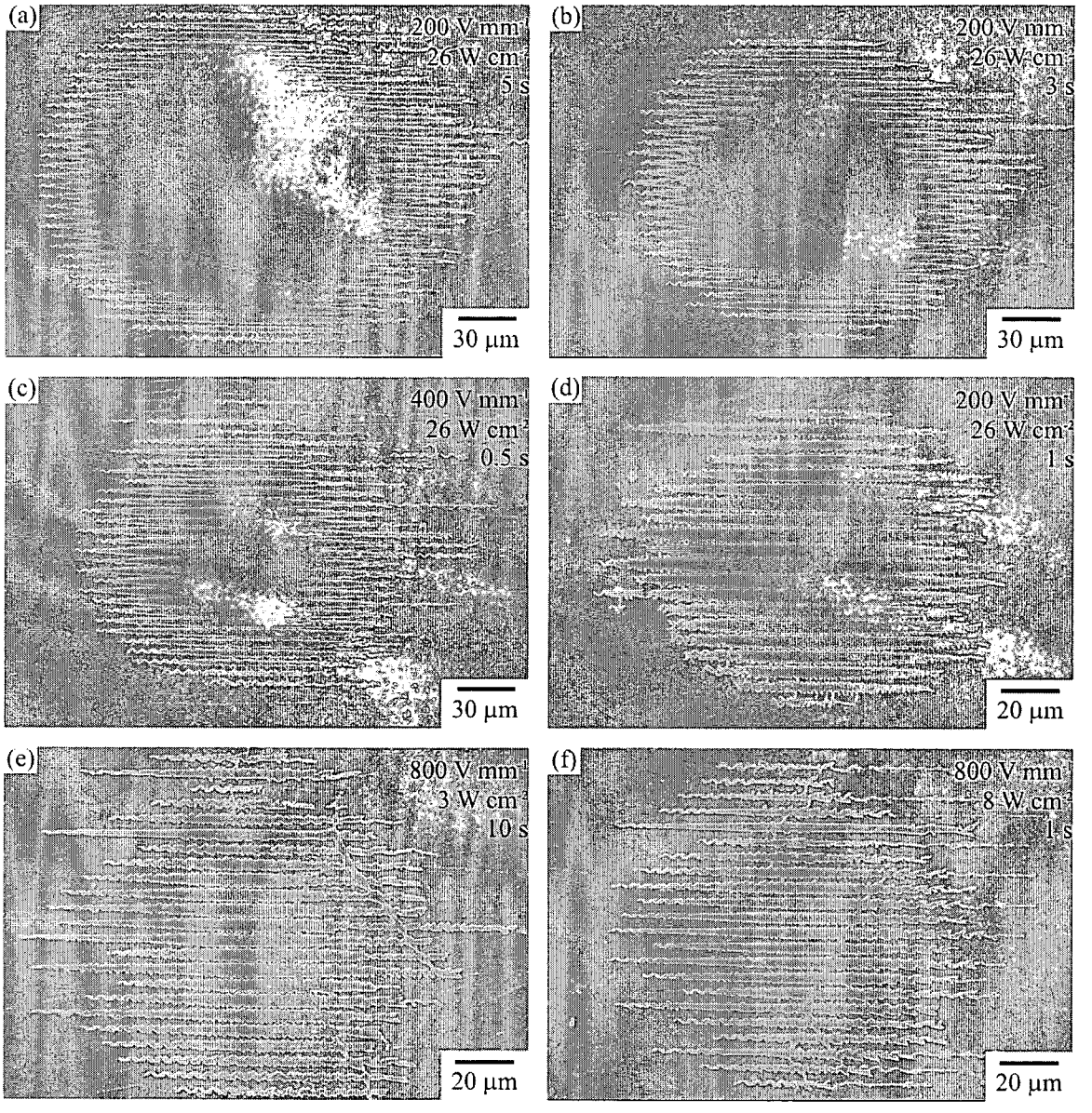

Figure 4: Domain pattern produced in 5-mol% MgO-doped congruently melting lithium niobate using light of wavelength $\lambda = 514.5$ nm with illumination period $\Lambda = 5.25$ μm. The exposure conditions used appear at the top-right corner of each image.

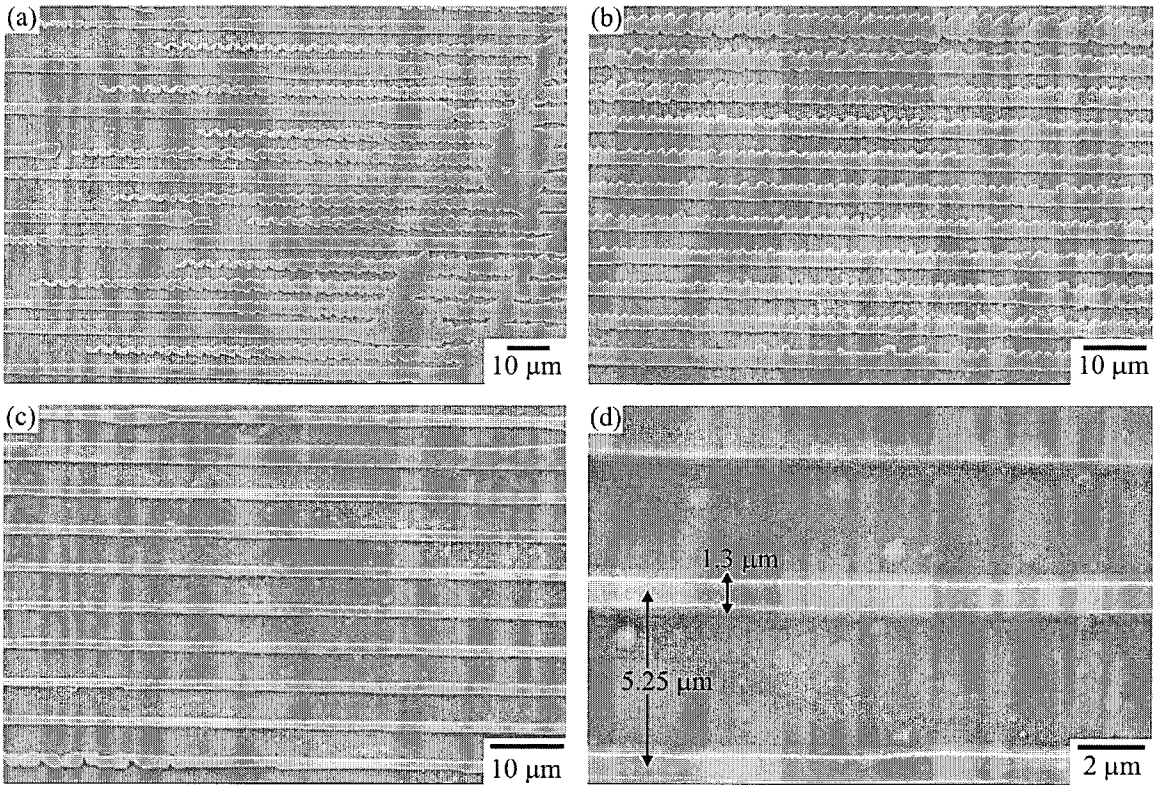

Figure 5: Domain pattern produced in 5-mol% MgO-doped congruently melting lithium niobate by scanning light of wavelength $\lambda = 514.5$ nm with illumination period $\Lambda = 5.25$ μm. The spot was scanned along the x-axis of the crystal, with an intensity $I = 20$ W cm^{-2} and electric field $E = 1200$ V mm^{-1}. (a), (b) Over-exposure leads to merging of adjacent domains and jagged edges. (c), (d) Improved exposure conditions with less spreading results in a high-quality periodic grating.