Appl Phys A (2010) 101: 313–317
DOI 10.1007/s00339-010-5823-9

Applied Physics A
Materials Science & Processing

Threshold fluence for domain reversal directly induced by femtosecond laser in lithium niobate

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Received: 29 October 2009 / Accepted: 10 May 2010 / Published online: 16 June 2010

Abstract Fabricating domain reversal directly induced by femtosecond laser is a novel and promising method to induce micron-period or even submicron-period inverted domain structure for it averts the domain spreading and mergence which is hard to avoid by traditional electric-field poling method. In this paper, the domain reversal process in lithium niobate crystal by irradiation of femtosecond pulses whose spatial and temporal distributions are taken into consideration is numerically simulated in the framework of Fahy's model. The simulation results manifest the domain inversion window theory and predict the threshold reversal fluence. The experiment to form domain reversal via direct illumination with femtosecond laser in Y-cut lithium niobate samples was conducted at room temperature. The multi-ring-like structures on the processed samples tally with the inversion window theory and the calculated threshold reversal fluence is well within the scope obtained by simulation, which serves as a corroborative evidence to prove the domain reversals can be formed by direct irradiation with femtosecond laser in lithium niobate.

1 Introduction

Forming domain reversal in ferroelectric crystal is vital for a wide range of application: quasi-phase-matched (QPM) nonlinear devices, switching and deflection devices and lens [1] based on electro-optic effect such as solc-filter and Q-switch, photonic bandgap structures, soliton formation [2, 3], and piezoelectric devices. Lithium niobate ($LiNbO_3$), regarded as silicon in nonlinear optics, is an ideal ferroelectric material for domain reversal as periodically poled lithium niobate (PPLN) for its excellent acousto-optic, electro-optic, elastic-optic, piezoelectric, pyroelectric, nonlinear optical, and optical waveguiding properties, and so on [4–6].

In recent decades, there emerge various methods to fabricate periodically poled inverted domain structure in lithium niobate crystal such as LiO_2 out-diffusion [7], proton-exchange [8], SiO_2-coating and heat treatment [9], Ti-indiffusion [10], and electron beam irradiation [11]. But these methods have not been employed broadly for the shallow reversal region or the demanding for certain temperature. The preferred and widespread domain engineering method is electric-field poling which is relatively more reliable and can fabricate bulk periodic structures at room temperature [12]. Nevertheless, the period of the PPLN produced by this method can just be reduced to approximately a few microns scale due to the unwanted domain broadening and even domain mergence [13] which obstruct its applications in first-order QPM nonlinear processes at UV wavelengths and electro-optic Bragg grating or photonic crystal structure requiring for submicron domain periods.

In 1994, reversal of ferroelectric domains by ultrashort optical pulses is suggested by Fahy et al. [14]. Since then, domain formation illuminated by ultraviolet laser pulse has been verified [15, 16]. Recently, a novel method to fabricate domain reversal structure in ferroelectric crystal by infrared femtosecond laser has been proposed. Since femtosecond laser can be focused to micrometer scale with high energy, domain inversion with micron-period or even submicron-period can be realized through femtosecond laser irradiation

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and there is no need to worry about the troublesome domain expanding and merging for such short-term interaction [17].

In this paper, a modified model in which an electric force term is added as well as the pulse distributions are detailed in both time domain and space domain is applied to imitate the course of domain orientation switch in $LiNbO_3$ primarily based on Fahy's work. Taking the sample characteristics and experimental fact into account synthetically, the predicted threshold fluence for domain reversal is proposed. At room temperature, an amplified Ti:sapphire femtosecond laser system was employed to induce inverted domain structures on the surfaces of Y-cut lithium niobate crystals. The multiring-like structures on the processed samples match well with the inversion window theory and the calculated threshold fluence of each reversal ring coincides with the numerically predictive range. The good agreement of the experimental results with the theoretical simulation acts as a corroborative evidence to identify the periodic structures as domain reversal.

2 Numerical simulations

As proposed by Fahy et al. originally [14], a simplified mode of a 30×30 two-dimensional oscillators array in X–Y plane (perpendicular to the Z-axis of the crystal) was adopted to simulate the process of the structural alteration of domain in lithium niobate crystal injected by a femtosecond laser pulse. In this mode, the librations of the Li-ions along the Z-axis are represented by the vibrations of the harmonically coupled and damped oscillators. Only considering about the nearest neighbor coupling with a coupling spring constant k since the long-range coupling does not affect our conclusion qualitatively [14], the motion equation of the system can be described as

$$\dot{v}_{i,j} = -4z_{i,j}^3 + 2az_{i,j} + k(z_{i-1,j} + z_{i+1,j} + z_{i,j-1} + z_{i,j+1} - 4z_{i,j}) - \gamma v_{i,j} + \eta + \frac{e}{m}E_{i,j} \tag{1}$$

in which, the subscripts are used to identify the oscillator in the array, $v_{i,j} = \dot{z}_{i,j}$ where $z_{i,j}$ is the relative position of the certain oscillator from the center of the two equilibrium points ($z_{\min\pm} = \pm\sqrt{a_1/2a_2}$) which are the minima of the anharmonic double-well potential formed by oxygen-plane $U(z) = -a_1z^2 + a_2z^4$ (z is the amplitude of the oscillator and a_1 as well as a_2 are two parameters determined by the nature of the crystal), k/a is given as 20 in reference to [14, 18, 19], the damping constant is signified by γ and a random force term η is simplified to be proportional to $\gamma k_B T$ in connection with thermal motion [20].

The last term in (1), in which e and m are the charge quantity and the mass of Li^+, respectively, shows the action of laser pulse which can be formulated as follows after nondimensionalization:

$$E_{i,j} = E_0 \exp\left[-\left(i^2 + j^2\right)/w_0^2\right] \exp\left[-b(t - t_0)^2\right] \times \cos(\omega_0 t - \varphi) \tag{2}$$

where E_0 is the peak value of electric field amplitude, w_0 is the radius of the laser waist spot, and the first exp-item indicates spatial distribution. The temporal part is represented by the rest of the items of (2) in which constant b shows the decay in time domain, ω_0 is the carrier envelop phase.

The Li-ions stabilized at the positive equilibrium point before incidence of pulse. If oscillators gain enough energy from the pulse to climb over the potential barrier at the tail of pulse, the system will be stable at another equilibrium position, which symbolizes the occurrence of the domain inversion. The simulation work is established on this thought and all the parameters are set in the light of the samples and femtosecond laser in the following experiment. In regard to the $LiNbO_3$ sample used in experiment, the parameters a_1 and a_2 are computed to be 1.87×10^{-19} J/nm^2 and 1.87×10^{-17} J/nm^4 separately and $k = 7000$ eV [21] as well as $\gamma = 1$ ps [22]. The wavelength and the duration of the pulse are 800 nm and 80 fs, respectively, in terms of the experimental reality. Thus, $b = 4 \times 10^{26}$ s^{-2} and the carrier envelope phase was set at $\pi/2$ to make the best usage of the pulse energy. T is set at 293 K since the experiment was carried on at room temperature.

The imitation work shows the stable position at the tail of the pulse turns to be positive and negative alternatively with the increasing of the electric field amplitude and the differences between the two shifts of the equilibrium position in succession are not the same, which satisfied the inversion window theory proposed by the work [17]. Growing the value of E_0 gradually and removing the unstable reversal range and the scope of single-shot ablation which is close to 4 J/cm^2 obtained by another experiment on the samples from the same company [23], it is achieved that the domain reversal is generated at certain fluence range illumination and the domain reversal fluence is within the range from 0.5 J/cm^2 to 4 J/cm^2, which has also been affirmed by the experimental conclusion.

3 Experimental

Experiments were conducted to verify the theoretical modeling predication. An amplified Ti:sapphire femtosecond laser beam with the wavelength of 800 nm and the duration of 80 fs at 1-kHz repetition rate was employed to focus on the surface of a Y-cut $20 \times 10 \times 1$ mm congruent lithium niobate crystal through an optical lens with a focal length of 150 mm. The pulse energy (from 30 μJ to 60 μJ, under the damage threshold) and polarization (in Z direction) were adjusted via a combination of half-wave plate followed by a

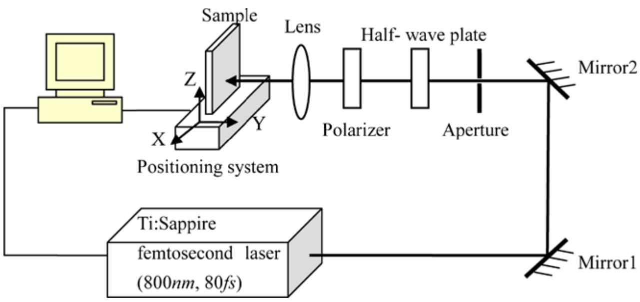

Fig. 1 Experimental setup for domain inversion induced by femtosecond laser in lithium niobate crystal

polarizer. The sample was positioned on a stepping-motor-controlled stage in the way that its X–Z-plane surface was normal to the incident direction of laser beam. The written repetition of laser and the translation speed of stepping-motor-controlled stage where the sample is fixed are set to be 10 Hz and 4 mm/s, respectively, to ensure it single-pulse action. Since the pulse duration is dozens of femtoseconds and the spot size is a few micrometers, the influence of movement can be neglected. A simplified picture of experimental setup is illustrated in Fig. 1.

4 Results and discussions

After hydrofluoric acid etching, multiring-like structures have been observed from a microscope through 400 times amplification, as shown in Fig. 2. The bright rings, illustrated as the white rings in the diagrammatic sketch Fig. 2(d), are recognized as the inverted regions for the dark circles and dark rings share the similar transmittance of light with the zones out of the radiation of femtosecond laser after hydrofluoric acid corrosion and the surface is sensitive to the HF acid to visualize the multiring-like morphological features. The appearances of inverted and noninverted regions in turns are in great accordance to the simulation results.

Comparing the diameters of the circles and rings in Fig. 2, it is obvious that the spot size become larger as the incident energy raised from 30 μJ to 60 μJ, beyond which, either no inversion occurred or perceptible ablation was detected. To get the threshold fluence to induce the domain reversal for each inverted area generated by different incident energy, the radius of each inverted ring has been measured. Since the dark rings (noninverted region) are too thin to be gauged precisely, the average radii have been noted as r_2, for the radii were marked by r_1, r_2, and r_3 from inner to outer as illustrated in Fig. 2(d). Seeing that the size of the waist radius cannot be obtained directly, the following data processing method was utilized to gain the threshold fluence of every inverted domain ring.

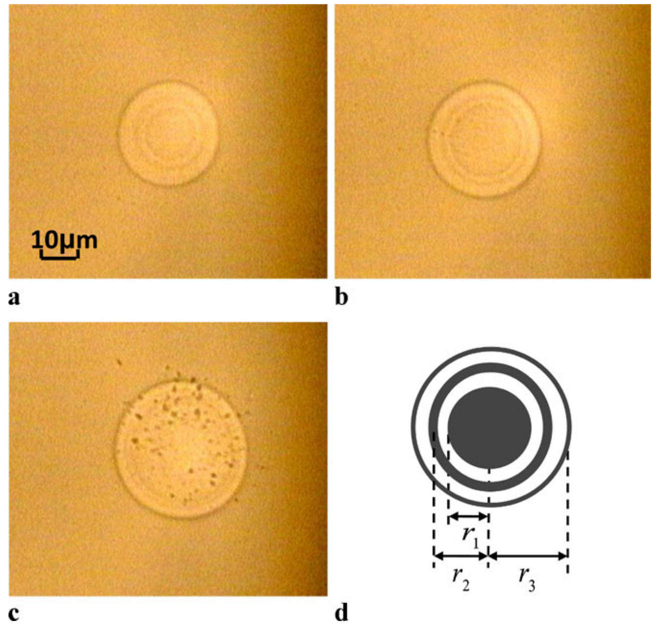

Fig. 2 Morphological features of the X–Z plane surface of $LiNbO_3$ irradiated by a Z-direction polarized femtosecond laser pulse with the energy (**a**) 30 μJ, (**b**) 40 μJ and (**c**) 60 μJ after hydrofluoric acid etching and the schematic diagram (**d**)

Similar with (2), the electric field of the laser pulse can be written as

$$E(r,t) = E_0 \exp\left(-\frac{r^2}{w_0^2}\right) \exp\left(-bt^2\right) \cos(\omega_0 t - \varphi) \tag{3}$$

Thus, the energy per unit area is given by

$$F = \int_{-\infty}^{+\infty} \varepsilon E^2(r,t) v_\varphi \, dt \tag{4}$$

in which, permittivity in the air ε approaches to the dielectric constant in vacuum ε_0 and the phase velocity v_φ can be approximated to the speed of light. Substituting the wavelength and the pulse width into the above formula, it turns to be

$$F = 3.01 \times 10^{-14} \varepsilon_0 c E_0^2 \exp\left(-\frac{2r^2}{w_0^2}\right) \tag{5}$$

Moreover, the single pulse energy can be expressed as

$$W = \int_0^{+\infty} \int_0^{2\pi} F r \, dr \, d\theta = \frac{\pi}{2} \times 3.01 \times 10^{-14} \varepsilon_0 c E_0^2 w_0^2 \tag{6}$$

Here, use W to represent energy in order to show a distinction with electric field. Therefore, the relationship between fluence and pulse energy is

$$F(r) = \frac{2\exp(-2r^2/w_0^2)}{\pi w_0^2} W \tag{7}$$

Table 1 Square differences between the radii of the corresponding rings of spots in Fig. 2

W_A (μJ)	W_B (μJ)	$r_{B1}^2 - r_{A1}^2$ (μm^2)	$r_{B2}^2 - r_{A2}^2$ (μm^2)	$r_{B3}^2 - r_{A3}^2$ (μm^2)	Average $r_B^2 - r_A^2$ (μm^2)
30	40	59.28	61.99	60.88	60.72
30	60	151.0	152.6	153.9	152.5
40	60	91.73	90.59	92.99	91.77

For the two spots A and B irradiated by two pulses with different energies W_A and W_B, respectively, if the two rings on these two spots have the same fluence, the square difference between the two radii of the rings satisfies

$$r_B^2 - r_A^2 = -\frac{w_0^2}{2} \ln\left(\frac{W_A}{W_B}\right) \tag{8}$$

That means for the certain waist radius the square difference between the two radii is just related to the values of incident pulse energies and independent of the concrete value of fluence if they have the same energy per unit area. After calculation and comparison, we found the square differences between the radii of the corresponding rings of spots in Fig. 2 fluctuated slightly (mainly caused by the measurement errors) as shown in Table 1. That is to say, domain inversion just occurs under certain fluence illumination regardless of the incident energy. Thus, with the enlargement of incident energy, the spot size grows to ensure irradiation with certain fluence.

Curve fitting of the data in Table 1 according to (8) enables us to gain the waist radius $w_0 = 21$ μm. Moreover, substituting the value of waist radius back into (7), the threshold fluence for each inverted region has been achieved. The reversal can be induced as the fluence is beyond 1.8 J/cm^2 and collapses with the fluence larger than 3.6 J/cm^2. There exists a thin noninverted region when the incident pulse fluence is nearby 2.9 J/cm^2, confirming to the simulation conclusion that reversal must be induced at certain ranges of pulse fluence. The reversal fluence scopes are in favorable conformity to the simulation results on basis of the principle of domain reversal, which to a large degree verifies that domain inversion can be induced in lithium niobate by direct irradiation with femtosecond laser.

5 Conclusion

An improved Fahy's model is proposed to simulate the inversion process in lithium niobate sample via femtosecond laser radiation and the parameters were set in accordance with experimental reality. The numerical imitation results demonstrate inversion and noninversion of domain occurs alternatively with the growing of incident electric field amplitude. The domain reversal just happens under specific ranges of fluence illumination and domain reversal fluence is within the range from 0.5 J/cm^2 to 4 J/cm^2. Experiments were conducted at room temperature to form domain switching in Y-cut lithium niobate crystals by irradiation with femtosecond laser straightly. The surfaces of samples show multiring-like structures after acid etching. The data processing work of radii of all inverted rings induced by the pulses with different energies indicates that the spot size increases with the enlargement of incident energy and domain reversal just occurs under certain fluence illumination irrespective of the incident energy. The calculated reversal fluence is from 1.8 J/cm^2 to 3.6 J/cm^2 except for a narrow noninverted scope nearby 2.9 J/cm^2, which agrees well with the simulated prediction. The wonderful fitting of the experimental results with the theoretical modeling corroborates that domain reversal can be fabricated via direct irradiation of femtosecond laser to a great extent.

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