Formation of domain reversal by direct irradiation with femtosecond laser in lithium niobate

Haisheng Zhu (朱海生), Xianfeng Chen (陈险峰)*, Hongyun Chen (陈洪云), and Xuewei Deng (邓学伟)

Department of Physics, the State Key Laboratory on Fiber Optic Local Area Communication Networks and Advanced Optical Communication Systems, Shanghai Jiao Tong University, Shanghai 200240

**E-mail: xfchen@sjtu.edu.cn*

Received June 16, 2008

We propose that domain inversion can be directly induced by femtosecond laser both theoretically and experimentally, which opens a path to achieve three-dimensional (3D) nonlinear crystal with a period in sub-micron-scale. A simulation of domain inversion is modeled by considering the temporal distribution of femtosecond pulses. The calculation results clarify that the domain inversions can happen within or after the interaction with the laser pulse, and the response time of domain inversion is in the picosecond level depending on the intensity and the materials. The domain reversal windows of lithium niobate by femtosecond laser are observed which agrees with theoretical predictions qualitatively.

OCIS codes: 320.0320, 160.0160, 310.0310.

doi: 10.3788/COL20090702.0169.

Periodically poled lithium niobate (PPLN), as a manual nonlinear optical crystal, has been widely used for frequency conversion in the past decade. In order to obtain short wavelength coherent optical sources by second harmonic generation (SHG) or sum frequency generation, micrometer scale periodic domain structure is necessary, which is out of the ability of conventional room temperature electrical poling techniques. As the technique of the femtosecond ultrafast laser develops, interactions between femtosecond laser and various materials have drawn more and more attentions. Femtosecond laser shows unique potential applications in micro-machining[1,2], optical storage[3], and optical processing[4,5]. As we know, femtosecond laser can be focused to micrometer scale, so if it can induce domain inversion in ferroelectrics crystals, domain inversion with micron-period can be realized. On the other hand, because the domain inversion happens under certain intensity, and the laser can be focused inside the crystal, it is expected that three-dimensional (3D) domain inversion can be achieved. In 1994, Fahy *et al.* suggested that reversal of ferroelectric domains can be directly achieved by ultrafast laser pulses[6], and recently optical poling by short ultraviolet (UV) pulses has been demonstrated[7,8]. Here we use a more detailed model to propose a method for domain inversion by femtosecond laser with the wavelength of 800 nm, the mechanism of which is different from Refs. [7,8].

Different models have been used to study ferroelectric materials driven by ultrafast optical pulses. As mentioned above, a simple model from Fahy *et al.* indicates properties of domain inversion subjected to a high-intensity optical pulse[6]. Another one from Montakhab *et al.* investigates the behavior of domain walls[9]. In this letter, followed by their works, we theoretically simulate the domain inversion process considering spatial and temporal distributions of femtosecond pulses with an oscillating electric field, in which ion accelerating process is studied. Different from the previous works, our simulations can provide the information about the energy threshold of domain inversion as functions of crystal parameters.

In the x-y plane, a model of an $n \times n$ oscillator array is considered, in which the oscillators, representing the lithium ions of lithium niobate, are harmonically coupled and damped[10]. Only the nearest neighbor coupling with a coupling spring constant k and an anharmonic double-well potential $u(z) = -az^2 + bz^4$ (z is the amplitude of the oscillator, a and b are coefficients) are taken into account[6,11]. When the laser pulse gives the oscillator enough energy to climb over the potential barrier, the system will be stable at another equilibrium position, then the inversion happens[12]. To start the oscillation of the ion oscillators, the polarization direction of the laser pulse should be parallel to the c axis (spontaneous polarization axis) of the single crystal congruent lithium niobate. The electric field of the laser pulse acting on the crystal is described as follows (generally, we choose Gaussian distribution):

$$E_{ij,0} = \frac{A}{u_0} \exp\left[\frac{-\left(i^2+j^2\right)}{u_0^2}\right]\Bigg|_{t_0}$$

$$= E_0 \exp\left[-B\left(i^2+j^2\right)\right], \qquad (1)$$

where $E_{ij,0}$ is the electric field amplitude at the peak value of the laser pulse at (i, j) in the array[13], u_0 is the radius of the laser waist spot, and $B = 1/u_0^2$ is the modulus decay of laser spot. Equation (1) describes the electric field spatially, which is also applied in Montakhab's model[9]. In order to investigate the influence of laser pulse parameters (response time, threshold, and duration, etc.), the temporal term should be considered. So we have

$$E_{ij}(t) = E_{ij,0} \exp[-\alpha(t-t_c)^2] \cos(w_0 t - \varphi), \qquad (2)$$

where α is the modulus decay of the laser pulse in temporal distribution, t_c is a constant, w_0 is the angular frequency of the electric field, and φ is the carrier envelop

phase (CEP).

When the oscillators are driven by a laser pulse, given by Eq. (2), the motion equation of the oscillators can be written as

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

where $v_{ij} = \dot{z}_{ij}$, (i, j) represents the positions of the oscillators in the array, $k/a = 20$ is given[14,15], the damping constant γ is of the order of $\sqrt{a}/10$ or larger[6,16], and η is a random force term equal to $\gamma k_{\mathrm{B}}T$ for simplification[17]. The last term on the right hand side of Eq. (3) is the laser pulse driven term, where $q = e$ for Li^+ and m is the mass of Li^+.

Since the coercive field in lithium niobate is about 21 kV/mm in room temperature, the parameters a and b in double-well potential $u(z) = -az^2 + bz^4$ can be found approximately as $a \approx 2 \times 10^{-22}$ (J/nm^2) and $b \approx 2 \times 10^{-24}$ (J/nm^4). In our calculation, all units are set equal to 1 and as a consequence all quantities are dimensionless. We set $a \in [1, 3]$ and $a = b$, which basically agrees with the physical situation. The laser frequency is 0.375×10^{15} Hz for 800-nm wavelength (time unit in the model is of order femtosecond). We define the datum time as t_1, which satisfies $\exp[-\alpha(t_1 - t_c)^2] = 0.001$. At $t = t_1$, all the oscillators are at the positive minimum of the double-well potential and the periodic boundary condition is used in this model[18]. Simulations under various conditions have been done.

A typical result among those simulations is shown in Fig. 1 with $\gamma = 0.4$, $k = 20$, $a = 1$, and $\eta = 0.1$. Figure 1 depicts the final average value of z of a 30×30 oscillator system for various values of E_0. The curve has a sharp turn at the domain inversion threshold, and becomes stable until the back-switch happens.

Domain inversion and back-switch will appear alternately as the value of E_0 grows. Domain inversion window is defined here as a continuous laser pulse energy range in which domain inversion can be excited by corresponding energy. As Fig. 1 suggests, the domain inversion windows have different sizes, and larger window appears when the electric field E_0 increases. The first domain inversion window requires low electric field E_0 and its width is so small that the inversion is not stable, which indicates that a little change of electric field E_0 will switch back the domain inversion. As for higher laser pulse energy, the width of domain inversion window becomes broader. At the boundary of the domain inversion window, the change of the electric field of the laser pulse has a great effect on the oscillators, so that a little change of the value of E_0 will cause the behavior of the oscillators totally different.

In Fahy's model, laser pulse's effect on the crystal is simplified as giving the oscillators an impulsive velocity, and the domain inversion happens during the relaxation time. In our simulations, we verify their assumption that permanent domain inversion has been achieved near the tail of the pulse, which means the response time is as a magnitude of picosecond. Figure 2 describes the behavior of the central oscillator in the 30×30 array irradiated by a laser pulse with a duration of 40 fs and an energy in the domain inversion window. When the laser pulse has passed through, the domain is not switched again. At the beginning, the electrical amplitude of laser pulse plays a dominant role, forcing the oscillators to vibrate at the same frequency. Then, when the peak of the laser pulse reaches the specimen, the influence of neighbor oscillators and the laser pulse both control the behavior of the oscillators, which leads the vibration to an irregular one during this period, as shown in Fig. 2. Because the coupling spring constant and the damping constant are settled, when the laser pulse has passed through, the Li ion, with a slight vibration, will finally become stable in the equilibrium state of domain inversion after sub-picosecond scale. As we can see, the domain reversal will not be switched again and the domain inversion by femtosecond laser is a permanent one.

Our simulation also shows the evolving process of the whole 30×30 oscillator array. At first, the electrical amplitude of pulse plays a dominate role. When the laser pulse has peaked, the vibrations of 30×30 oscillator array become irregular owing to the interactions between the oscillators.

In our simulations, the parameters of oscillators for lithium niobate are uncertain and depend on the crystal itself and the ambient circumstance. In order to investigate the influence of parameters of the oscillators, we calculated the domain inversion threshold as functions of the damping constant, the coupling spring constant, and the ambient temperature.

Figure 3(a) shows the domain inversion threshold

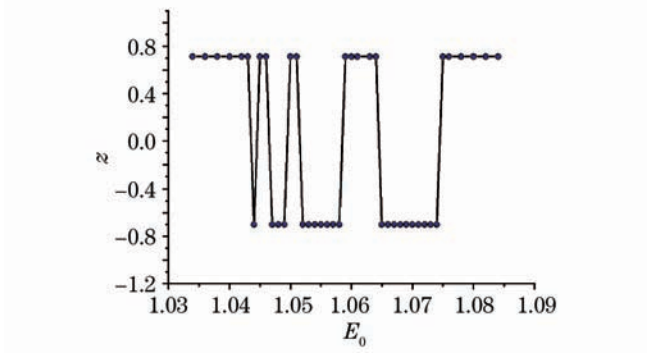

Fig. 1. Several domain inversions shown as final average z versus electric field E_0 for a 30×30 oscillator system evolving. The parameters are $\gamma = 0.4$, $k = 20$, $a = 1$, and $\eta = 0.1$.

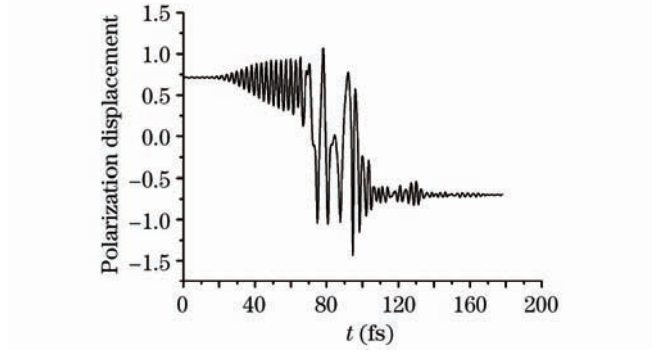

Fig. 2. Behavior of the central oscillator in the 30×30 array irradiated by a laser pulse with a width of 40 fs.

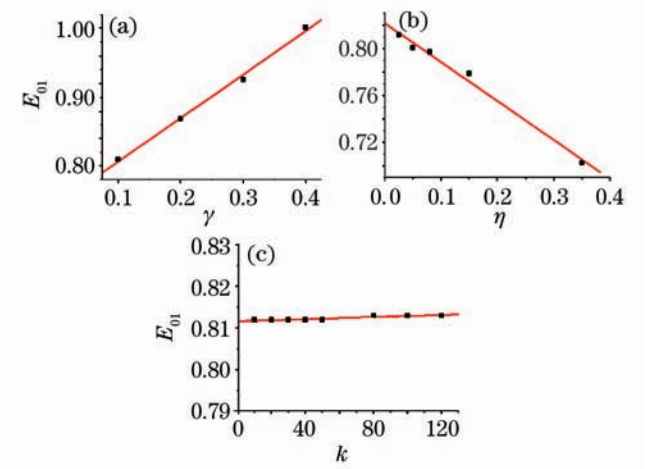

Fig. 3. (a) Domain inversion threshold E_{01} versus damping constant γ ($a = 1$, $k = 20$, $\eta = 0.25\gamma$); (b) domain inversion threshold E_{01} versus coupling spring constant k ($a = 1$, $\gamma = 0.1$, $\eta = 0.025$); (c) domain inversion threshold E_{01} versus force term η which is proportional to the ambient temperature ($a = 1$, $k = 20$, $\gamma = 0.1$).

versus the damping constant γ. E_{01} is the lower threshold of domain inversion window, and the width of domain inversion is E_0-window. As γ grows, the values of E_{01} and E_0-window grow too. Obviously, larger γ requires more energy for oscillators to climb the potential barrier, which results in the higher E_{01}. Besides, larger γ also requires more energy for switch-back, which results in broader E_0-window. Note that if γ is too small, the oscillators need a long time to become stable, and it means the response time of domain reversal becomes longer.

The growth of coupling spring constant k will increase the values of E_{01}, and its effect on the behavior of oscillators is rather small, as shown in Fig. 3(b). The reason is that k is a recovery factor of a spring and it relates to frequency instead of energy. Temperature also influences the domain inversion, higher temperature, which is proportional to η, requires lower laser energy for domain inversion, as shown in Fig. 3(c). It should be mentioned that the selected temperature should be lower than the paraelectric transition temperature of the crystal.

To verify these predictions, an initiative experiment has been done. A y-cut lithium niobate crystal was used in the experiment. An amplified Ti:sapphire femtosecond laser with the wavelength of 800 nm and the duration of 80 fs was employed. The repetition rate of the laser was 1 kHz. Figure 4 shows the morphological features of the lithium niobate sample irradiated by femtosecond laser with a power lower than damage threshold and followed by HF acid etching. At first, the polarization of laser was set to be parallel to the z-axis of the crystal. When the laser power exceeds a certain threshold, domain reversal, shown as a circle, was observed on the surface of the y-face after HF acid etching. When the intensity was futher increased, the circle became larger. The observations are agreeable qualitatively with our calculations above. In the experiments, the laser power of 40 and 60 mW were used. Because the response of surface to the HF acid is distinct, it is shown that domain inversion happened within the circle. Then, the polarization of laser was turned along the x-axis of the crystal, with the same

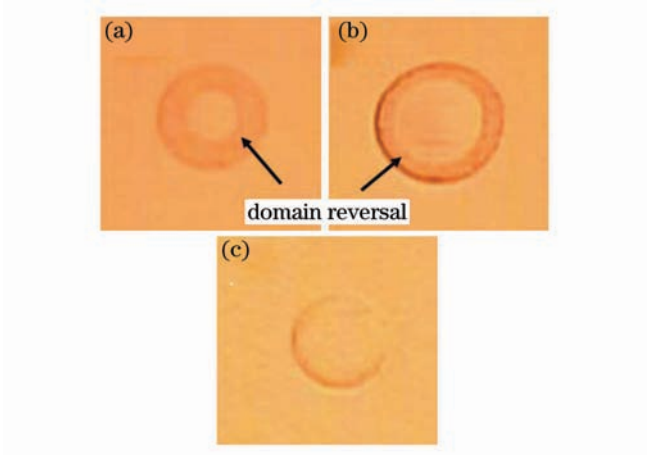

Fig. 4. Morphological features of the lithium niobate sample irradiated by laser pulse with wavelength of 800 nm, duration of about 80 fs, and powers (polarization) of: (a) 40 mW (z); (b) 60 mW (z); (c) 40 mW (x).

power of 40 mW as that in Fig. 4(a), no domain reversal circle was observed, as shown in Fig. 4(c). More detailed experimental demonstrations are in process.

In summary, based on the improved classical oscillators model, we find that femtosecond laser pulse electric field can directly induce the domain inversion in lithium niobate crystal. The influence of parameters of the model on the domain inversion is studied. The decrease of the damping constant and the coupling spring constant, and the increase of temperature will reduce the domain inversion threshold power. The results of our experiments obviously agree with the predictions. These studies give useful information for realizing 3D short-period nonlinear crystals by choosing suitable parameters of the femtosecond laser pulse.

This work was supported by the National Natural Science Foundation of China (No. 10734080 and 10574092) and the National "973" Program of China (No. 2006CB806000).

References

1. F. Beinhorn, J. Ihlemann, P. Simon, G. Marowsky, B. Maisenhölder, J. Edlinger, D. Neuschäfer and D. Anselmetti, Appl. Surf. Sci. **138-139,** 107 (1999).
2. X. Ni, C. Wang, Y. Wu, L. Yang, W. Jia, and L. Chai, Chin. Opt. Lett. **4,** 225 (2006).
3. E. N. Glezer, M. Milosavljevic, L. Huang, R. J. Finlay, T.-H. Her, J. P. Callan, and E. Mazur, Opt. Lett. **21,** 2023 (1996).
4. H. Yang, J. Zhang, W. Yu, Y. J. Li, and Z. Y. Wei, Phys. Rev. E **65,** 016406 (2001).
5. T. Kondo, S. Matsuo, S. Juodkazis, and H. Misawa, Appl. Phys. Lett. **79,** 725 (2001).
6. S. Fahy and R. Merlin, Phys. Rev. Lett. **73,** 1122 (1994).
7. C. E. Valdivia, C. L. Sones, J. G. Scott, S. Mailis, R. W. Eason, D. A. Scrymgeour, V. Gopalan, T. Jungk, E. Soergel, and I. Clark, Appl. Phys. Lett. **86,** 022906 (2005).
8. I. T. Wellington, C. E. Valdivia, T. J. Sono, C. L. Sones, S. Mailis, and R. W. Eason, Appl. Surf. Sci. **253,** 4215 (2007).
9. A. Montakhab and J. Levy, Phys. Rev. E **56,** 6082

(1997).
10. S. L. Chaplot and K. R. Rao, J. Phys. C **13,** 747 (1980).
11. B. M. Fridin, *Photoferroelectrics* (in Chinese) D. Xiao, (trans.) (Science Press, Beijing, 1987) Chap.2, 3.
12. J. Li, in *Dielectric Medium Physics* (in Chinese) J. Fang and Z. Yin, (eds.) (Science Press, Beijing, 1989) Chap.5.
13. O. Svelto, *Principles of Lasers* (2nd edn.) (Plenum Press, New York, 1982) Chap.8.
14. J. A. Krumhansl and J. R. Schrieffer, Phys. Rev. B **11,** 3535 (1975).
15. G. Shirane, Rev. Mod. Phys. **46,** 437 (1974).
16. T. P. Dougherty, G. P. Wiederrecht, K. A. Nelson, M. H. Garrett, H. P. Jensen, and C. Warde, Science **258,** 770 (1992).
17. H. Risken, *The Fokker-Planck Equation* (2nd edn.) (Springer, Berlin, 1989) Chap.3.
18. H. Li, *Introduction to Dielectric Physics* (in Chinese) (Chengdu Science and Technology University Publishing Company, Chengdu, 1990) Chap.5.