

## OPTICAL CONTROL OF DOMAIN STRUCTURES IN LITHIUM TANTALATE CRYSTALS

A. R. Poghosyan\*

Institute for Physical Research, Ashtarak-2, 378410, Armenia

We present an alternative method for production of periodically patterned domain structures due to optical periodic poling, a technique involving the simultaneous application of combined electrical and optical fields: the electric field is applied via planar electrodes, while light is used to define those regions where domain inversion should occur. The optical poling route therefore offers a potentially simpler method, effectively eliminating the photolithographic patterning steps. Using interferometric methods, the periodicity of optically induced domain structures is a function of laser wavelength, and intersection angle of the two interfering beams, and hence is easily changed. The investigations of the mechanisms of optical control of domain structures in ferroelectrics, in particular the role of various internal field components, their origins and dynamic behavior following domain reversal are presented.

(Received July 10, 2003; accepted July 31, 2003)

*Keywords:* Ferroelectrics, Periodic poled domain structures, Lithium tantalate

### 1. Introduction

Quasi-phase matching by periodical inversion of ferroelectric domains can achieve high conversion efficiency for second harmonic generation. Intensive studies on fabrication, characterization and performance of second harmonic generation devices on periodically inverted ferroelectric domain LiTaO<sub>3</sub> crystals are taking place, but studies on basic material properties of LiTaO<sub>3</sub> in the domain inversion process are lacking. On the other hand all post-growth techniques for achieving periodic domain profiles require either specialist clean-room photolithographic fabrication of patterned electrodes, or lengthy thermal or electron-beam scanning processes. Recently the experiments demonstrating the advantages of direct optical control of ferroelectric domain profiles have been carried out [1, 2]. We have recently shown that it is possible to obtain bulk periodically poled LiNbO<sub>3</sub> single crystals with antiparallel ferroelectric domains, by direct electric field poling during growth process [3]. Here we present our findings and detailed analysis on time dependence of the ferroelectric coercive field after domain inversion of the LiTaO<sub>3</sub> crystal and report a simple room temperature technique for patterning the domain profile by the simultaneous application of optical and electric fields without the need for photolithographic processing.

### 2. Experimental results

LiTaO<sub>3</sub> crystals used in our experiments were 0.2 mm thick, z-cut optical grade wafers (x, y sides were 10 mm) of Czochralski grown material supplied by Mitsui Corporation, Japan. At room temperature LiTaO<sub>3</sub> is ferroelectric (crystal class *3m*, spontaneous polarization  $P_S = 50 \mu\text{C cm}^{-2}$ ). The direction of  $P_S$  is reversed easily by applying an electric field along Z-axis in excess of the coercive

---

\* Corresponding author: arp@ipr.sci.am

field  $E_C$ . The external field was applied to the crystal plate through liquid electrodes ( $\phi=5$  mm) and was ramp up from 2 kV at fixed rate (see Fig. 1).

The displacement current was monitored during the poling processes (Fig. 2). The external field at which the maximum displacement current occurs is defined as the switching field of the crystal. It is known that the switching fields in forward direction  $E_f$  (from “virgin” state of crystal to domain reversed state) and in reverse direction  $E_r$  (from domain reversed state to “virgin” state) are different, i.e. the ferroelectric hysteresis loop of  $\text{LiTaO}_3$  is asymmetrical [2]. Moreover it was found that  $E_f$  and  $E_r$  depend also on time between two successive poling operation (we define it as time interval  $\Delta t$ ) and light intensity [4]. In [5] it was shown that the asymmetry of switching electric fields depends on temperature and there is necessity of stabilization time after switching.

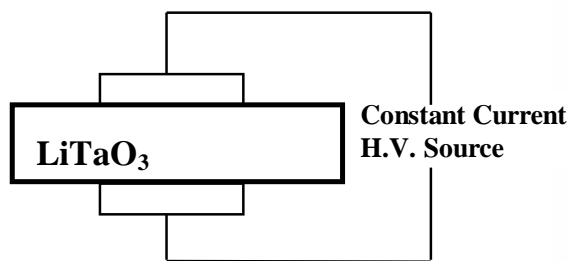


Fig. 1. The experimental setup.

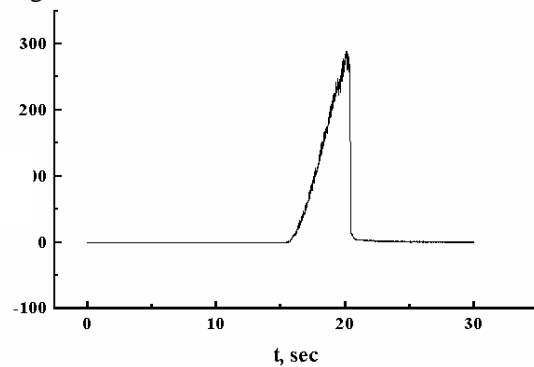


Fig. 2. The displacement current during the poling process.

In Fig.3 the switching electric field in forward direction vs time interval  $\Delta t$  measured in our experiments is presented.

One can see there is a strong dependence of switching electric field on time interval between two successive poling operations. Minimal time interval was 30 sec (that was defined by technical parameters) so we can not investigate the stabilization times and other phenomena near  $\Delta t = 0$ .

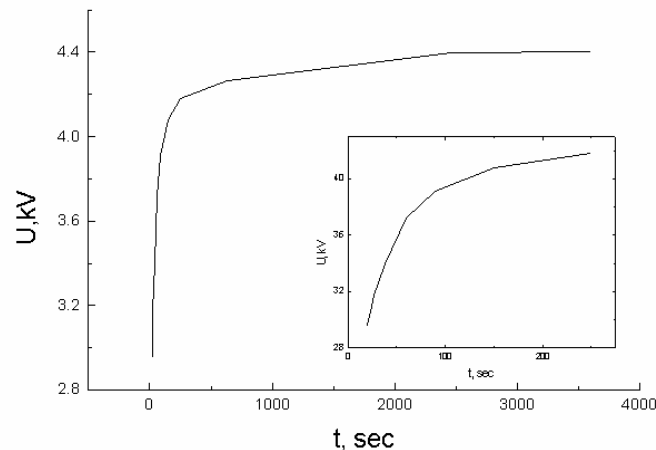


Fig. 3. Switching electric field vs time interval  $\Delta t$  in forward direction.

### 3. Model of poling processes

Now we consider in more detail the physical processes occurring during poling of ferroelectric crystals. In the single domain ferroelectric crystal with spontaneous polarization  $P_S$  there is an electric field  $P_S/\epsilon\epsilon_0$  (the depolarization field  $E_d$ ). It is clear from energetic reasons that this field has to be screened. It is known also that even in wide band dielectric the screening may be internal and of electronic nature. The availability of surface electronic levels may also give the essential contribution to the screening. The screening, reduces the free energy of a single domain crystal, and makes possible the

existence of single domain crystals. By cooling the crystal from phase transition temperature  $T_C$  up to room temperature there is a continued growth of polarization (and  $E_d$ ) and correspondingly it is necessary the increasing of space charge field  $E_s$  which has to screen  $E_d$ . Note that in various temperature intervals the screening is made by those carriers, which determine the conductivity of a crystal. In lithium tantalate, as it is known, the conductivity at  $T > 150$  °C has ionic character and at  $T < 150$  °C – electronic character. Thus a space charge field  $E_s$ , screening the depolarization field  $E_d$ , consists of two components: electronic  $E_e$  and ionic  $E_i$ .

So the total internal field in the crystal is:  $E_{int} = (E_d - E_s)$ . In steady state this field is equal to zero and the switching field is equal to the coercive field  $E_C$ . The screening space charge is concentrated in a surface layer of the crystal, the thickness of which is equal to a Debye length of a screening  $L_D = (\epsilon\epsilon_0 kT / e^2 n)^{1/2}$ , where  $n$  is the carrier concentration.

Let now consider the processes of poling and repoling in a crystal (Fig. 4).

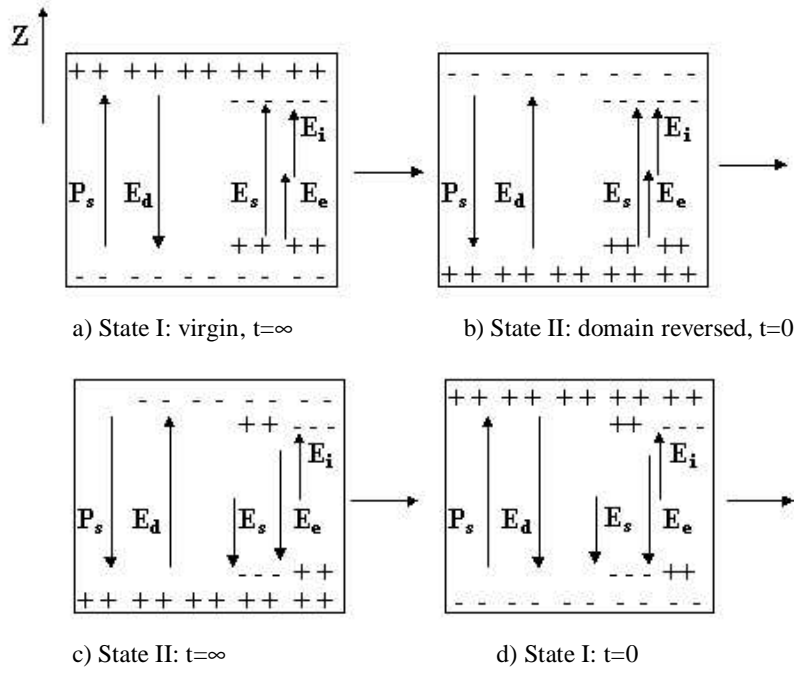


Fig. 4. Electric fields during consistent poling processes.

Fig. 4 (a) corresponds to virgin state of crystal when we can write for switching electric field in forward direction:

$$E_f(\infty) = E_c - E_{int} = E_c - (E_d - E_s) = E_c - E_d + E_e + E_i = E_c' + E_e + E_i = E_c^{ef} \quad (1)$$

Here  $E_c^{ef}$  is the effective coercive electric field of the crystal and  $E_c^{ef} \approx E_c$ . After switching in forward direction  $P_s$  and  $E_d$  change their direction but  $E_s$  at  $t = 0$  does not change and becomes antiparallel to the new polarization direction (and parallel to the new depolarization field direction). So we can write for switching electric field in reverse direction at  $t = 0$  (Fig. 4b):

$$E_r(t=0) = E_c - E_{int} = E_c - (E_d + E_s) = E_c - E_d - E_e - E_i = E_c' - E_e - E_i = (E_c^{ef} - 2E_e - 2E_i) \quad (2)$$

This state is not stable because there is a large internal field  $E_i = E_d + E_s$  in the crystal. So the space charge field  $E_s$  in this domain reversed state tends to realign antiparallel to the new depolarization field direction with time (note, if  $E_c^{ef} - 2E_e - 2E_i < 0$  then we need a time for stabilization of state II; we can define this time as time when  $E_r(t) = 0$ ). But this realignment is incomplete even after long time at room temperature because only electronic component of space charge field  $E_e$  may realign due to

electronic conductivity while ionic component stays stable (at room temperatures the ionic conductivity is “frozen”). So the internal field in this state is not equal to zero (below we more detailed consider why the electronic component can not completely screen the ionic field; but briefly we can say that the situation is very similar with the temperature fixing of holographic recording) and we have (see Fig. 4c):

$$E_r(\infty) = E_c - E_{int} = E_c - (E_d - E_s) = E_c - E_d + E_e - E_i = E'_c + E_e - E_i = (E_c^{ef} - 2E_i) \quad (3)$$

We can see that there is a large asymmetry for forward (1) and reverse (3) switching fields and it is equal to  $2E_i$ . It means we have large internal field in State II (there are some experimental evidences of the existence of a strong internal electric field after domain inversion <sup>4</sup>). After poling crystal from domain reverse state II back to the virgin state I we have (Fig.4d):

$$E_f(t=0) = E_c - E_{int} = E_c - (E_d + E_s) = E_c - E_d - E_e + E_i = E'_c - E_e + E_i = (E_c^{ef} - 2E_e) \quad (4)$$

The situation in the state I (at  $t=0$ ) is like the situation in the state II at  $t=0$ . In this state there is also the stabilization time (if  $E_c^{ef} - 2E_e < 0$ ), but in this case it have to be smaller. Then, after screening of depolarization field, we again obtain the crystal in virgin state (Fig.4a and eq. (1)).

From this phenomenological consideration of the poling processes we can write the joint expression for necessary switching electric fields in forward and reverse directions:

$$E_{f,r}(t) = E'_c + E_e (1 - 2\exp(-t/\tau_e)) + E_i (1 - 2\exp(-t/\tau_i)) = E_c^{ef} - 2E_e \exp(-t/\tau_e) - 2E_i \exp(-t/\tau_i) \quad (5)$$

where  $\tau_e$ ,  $\tau_i$  - relaxation times for electronic and ionic components of space charge electric field.

In (5) there is no difference between virgin and domain reversed states or between forward and reverse poling processes. This equation is correct if  $\tau_i$  is not very large. But, because  $\tau_i = \epsilon\epsilon_0/\sigma_i$  ( $\sigma_i$  - ionic conductivity of the crystal), we can use eq. (5) only when the crystal temperature  $T > 150$  °C. At lower temperatures due to “frozen” ionic conductivity we obtain different expressions for forward and reverse poling:

$$E_f(t) = E'_c + E_e (1 - 2\exp(-t/\tau_e)) + E_i = E_c^{ef} - 2E_e \exp(-t/\tau_e) \quad (6)$$

$$E_r(t) = E'_c + E_e (1 - 2\exp(-t/\tau_e)) - E_i = E_c^{ef} - 2E_e \exp(-t/\tau_e) - 2E_i \quad (7)$$

Note that virgin and domain reversed states are distinguished only by their “history”. If we heat the crystal in domain reversed state up to 150 °C and activate the ionic conductivity then the domain reversed state is become the virgin state and contrary.

Now we will try to calculate the parameters of the eq. (6) and (7) using experimental data. From Fig. 3 and <sup>2,3</sup> we can obtain:  $E_f = 210$  kV/cm,  $E_r = 130$  kV/cm,  $E_i = 40$  kV/cm.

The approximation of an experimental dependence  $E_f(t)$  on Fig. 3 with the help of equation (6) shows, that the experimental curve is described by the equation which is similar (6), but have two exponents (we consider yet not the weak dependence of  $E_f$  on  $t$  at  $t > 300$  s; this fact we'll consider below):

$$E_f(t) = E'_c + E_e^f (1 - 2\exp(-t/\tau_f)) + E_e^s (1 - 2\exp(-t/\tau_s)) + E_i \quad (8)$$

where  $E_e^f + E_e^s = E_e$ . The parameters' values obtained from Fig. 3 are equal to:  $E'_c = 50$  kV/cm,  $E_e = 120$  kV/cm,  $E_e^f = 90$  kV/cm,  $E_e^s = 30$  kV/cm,  $\tau_f = 6$  sec,  $\tau_s = 130$  s.

On Fig. 5 the dependencies of  $E_f(t)$  and  $E_r(t)$  calculated from eq. (8) are shown. You can see, that the equation (8) describes both the asymmetry of switching fields and asymmetry of stabilization times.

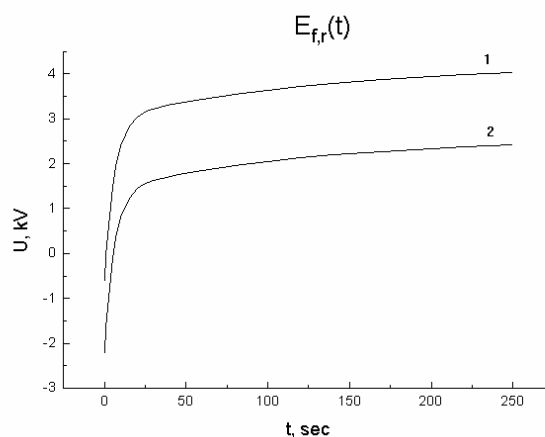


Fig. 5. The calculated dependencies of switching electric fields in forward (1) and reverse (2) directions on time interval between poling processes.

#### 4. Discussion and conclusion

Let's consider a physical sense of equation (8). Earlier we noted that  $E_i$  is the uncompensated part of an ionic component of a space charge electric field screening a depolarization electric field. We have named two other parameters  $E_f$  and  $E_s$  as fast and slow components of an electrical field screening the spontaneous polarization. The physical nature of a fast component is not so clear. As it was mentioned in [5] it may be a reorientation of dipole moments of point defects or it may be high conductivity in very high electric fields. In any case for clearing up of a nature of this component the additional investigations are necessary. As to a slow component this component has an only electronic nature and is conditioned by redistribution of electrons' space charge due to electronic conductivity. This supposition is confirmed by the fact that the relaxation time  $\tau_s$  rather well coincides with a Maxwell relaxation time  $\tau_m$  (direct measurement has shown, that dark conductivity for our samples is equal  $3 \cdot 10^{-13} \Omega^{-1} \text{cm}^{-1}$  and corresponds  $\tau_m = 120 \text{ sec}$ ). It means, that the relaxation time (and  $E_{f,r}$ ) depends also on light intensity because  $\tau_m = \epsilon \epsilon_0 / (\sigma_d + \sigma_{ph})$ , where  $\sigma_{ph}$  is a photoconductivity. Such dependence was observed in experiments [6] and also in our experiments. One practical conclusion also ensues from this. As it is seen from (8) the magnitude of a switching field in case of illumination of crystal in a defined time frame is higher than without illumination. Thereby for partial illumination of crystal the illuminated sites are poled at the higher values of an applied field than not illuminated. So there is a possibility of control of the domain structure with the help of light. Naturally for this it is necessary  $\sigma_{ph} > \sigma_d$ .

We shall return to a question why after poling the complete compensation of depolarization fields does not occur and there is an asymmetry of switching fields in forward and reverse directions. As we already marked, on Fig. 3 there is a weak dependence of  $E_f$  from  $t$  at  $t > 300 \text{ sec}$ ., i.e. the curve  $E(t)$  is not actually described completely by the exponent. In this area  $\tau_m$  begins to increase and the screening of depolarization field goes slower and slower, and in domain reversed state the complete compensation is not reached.

One of reasons of  $\tau_m$  increase may be a reduction of a charge carriers concentration in the crystal volume owing to a very large concentration of the carriers in a surface layer (the concentration of carriers in the  $L_D$  area should reach  $10^{18} \text{ cm}^{-3}$ ). And the concentration of carriers necessary for complete screening, naturally, depends on value of electric field, which should be screened ( in state II:  $(E_d + E_i)$ , in state I:  $(E_d - E_i)$ ). Thus, at identical concentration of carriers, the value of a noncompensated electric field in domain reversed state is higher than in virgin state, as results in asymmetry of switching fields.

Other possible reason may be diffusion current owing to a same large concentration of carriers in the  $L_D$  layer (similarly to a temperature fixing of the holograms). This process is essential in domain reversed state, where it is necessary to compensate much larger fields. As this effect works on the  $L_D$  length, it results in the increase of  $L_D$ , i.e. in the increasing of a noncompensated internal field in a crystal. However, unfortunately, at the present time the experimental data are not enough for an unequivocal conclusion about the mechanism of this process.

As we have mentioned above our experiments have shown the possibility of direct optical control of domain patterning in  $\text{LiTaO}_3$  crystals [6,7]. We used special z-cut crystals for obtaining a light interference pattern in the crystal volume (see Fig. 6). To optimize the fringe stability, and reduce Fresnel losses from additional optics, a biprism was polished directly onto Y-face. The apex thus formed bisected the incident UV laser beam, ensuring good spatial overlap beneath the electrode area. The polish angle was chosen so that the interference pattern formed had a periodicity that was both small enough to demonstrate the utility of the technique, but large enough to be easily visible under a polarizing optical microscope. The calculated periodicity within the crystal was  $5.2 \mu\text{m}$  for an angle  $1.5^\circ$ . However, due to polishing tolerances available, the inherent divergence of the incident laser beam, and the use of a weakly convergent cylindrical lens to help increase the incident UV irradiance, the measured periodicity obtained from optically frustrated poling here was  $\sim 6.3 \mu\text{m}$ .

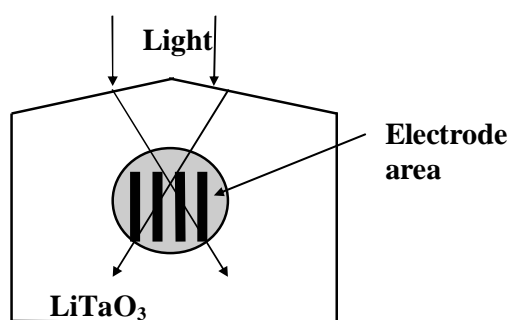


Fig. 6. Experimental setup for periodical poling using optical control.

Optical illumination was provided by UV lines (predominantly 364 and 351 nm) of an Ar ion laser. Using cylindrical optics the beam was launched into the wafer through the +Y face. In order to assess the effect of the UV illumination during poling process the resulting structure was examined under an electronic microscope after etching of Z-face of the crystal. After simultaneous applying of UV light and switching electric field for poling in the forward direction we have obtained periodically domain-inverted structure under the electrode area with  $\sim 6.3 \mu\text{m}$  period.

### Acknowledgements

The author would like to acknowledge the partial financial support for this work by ANSEF Award number PS 115.

### References

- [1] G. W. Ross, P. G. R. Smith, R. W. Eason, *Appl. Phys. Lett.* **71**, 309 (1997).
- [2] V. Gopalan, M. C. Gupta, *Appl. Phys. Lett.* **68**, 888 (1996).
- [3] I. A. Ghambaryan, R. Guo, R. K. Hovsepian, A. R. Poghosyan, E. S. Vardanyan, V. G. Lazaryan, *J. Optoelectron. Adv. Mater.* **5**(1), 61 (2003).
- [4] S. Chao, C. Hung, *Appl. Phys. Lett.* **69**, 3803 (1996).
- [5] V. Gopalan, T. E. Mitchell, *J. Appl. Phys.* **83**, 941 (1998).
- [6] P. T. Brown, G. W. Ross, R. W. Eason, A. R. Poghosyan, *Optics communications* **163**, 310 (1999).
- [7] A. R. Poghosyan, *Proceedings of SPIE* **3992**, 137 (2000).