TAILORING FERROELECTRIC DOMAIN CONFIGURATIONS FOR NONLINEAR OPTICAL DEVICES

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Abstract : Ferroelectric domain engineering became an attractive and perspective tool for development of a new generation of diverse nonlinear optical and acoustic converters. Conventional one-dimensional engineered domain structure represents an array of strip-like domains of a micrometer width. These domains should possess vertical domain walls through the crystal bulk of a few millimeters depth. In fabrication of patterned domain configurations by electrical poling technique, the most critical point is a domain broadening effect. There are several fundamental processes, which play a crucial role in fabrication of domain configurations, such as bulk domain nucleation, domain shapes and anisotropic domain walls propagation

Oblikovanje feroelektričnih domen pri izdelavi nelinearnih optičnih komponent

Ključne besede: optika nelinearna, KTP KTiOPO$_4$ kristali feroelektrični, naprave optike nelinearne, pretvorniki akustični, pretvorniki optični, inženirstvo, elektrooptika, piezoelektrika, piroelektrika, viri svetlobe koherentne, viri svetlobe vidne koherentne, viri svetlobe UV ultravijolične koherentne, viri svetlobe IR infrardeče koherentne

Izvleček : Inženiring feroelektričnih domen je postalo privlačno in obetajoče orodje za razvoj nove generacije različnih nelinearnih optičnih in akustičnih pretvornikov. Konvencionalna enodimenzionalna domenska struktura je sestavljena iz polja podolgovatih domen z mikrometrsko širino. Omenjene domene imajo navpične stene, ki se širijo skozi telo kristala v globino nekaj milimetrov. Pri izdelavi domenskih struktur z izbrano predlogo z metodo selektivne električne polarizacije je najbolj problematičen pojav razširitve domen. Pri tovrstnem oblikovanju domen je pomembnih kar nekaj osnovnih procesov .To so predvsem : nukleacija domen v telesu kristala, geometrija domen in anizotropno širjenje domenskih sten.

1. Introduction

Fabrication of engineered domain structures allows observing new physical properties of a ferroelectric crystal. The physical origin of this phenomenon is changes in a macroscopic symmetry of the domain-patterned ferroelectrics. Many well-known properties of ferroelectrics such as electrooptical, nonlinear optical, pyroelectric, piezoelectric, etc., which are determined by tensors of the odd ranks, may be changed by means of tailoring specific domain-engineered configurations /1/. In nonlinear optics the domain-engineered structures allow satisfying quasi-phase-matching (QPM) conditions /2/. This method enables to develop coherent light sources in UV, visible and infrared regions where compact and efficient lasers are unavailable.

In this paper the principles of ferroelectric domain engineering, peculiarities of the spontaneous polarization reversal, ferroelectric domains nucleation problem, and the domain propagation in ferroelectric materials are discussed. KTiOPO$_4$ (KTP) ferroelectric crystals have been chosen for this study, because of their wide application in nonlinear optical conversions. These crystals possess high optical nonlinearity, high optical damage resistance, and broad optical transparency.

2. Ferroelectric domain engineering

Ferroelectrics are primary ferroic (twinned) crystals where twins are distinct in the orientation of spontaneous polarization P_s. These electrical twins (ferroelectric domains) differ in the signs of every polar property of the first, third and any other odd rank tensors. Tensors of the third rank describe several important physical effects such as piezoelectric, electrooptic and nonlinear optical. In ferroic crystals twins are reoriented by external driving force that is an electric field for ferroelectrics. Therefore, external electric field reversing spontaneous polarization P_s reverses the signs of coefficients of the corresponding tensors of the odd rank. For instance, two adjacent domains with opposite direction of P_s have opposite signs of the piezoelectric coefficients. Evidently, application of the operating voltage

to this bi-domain ferroelectric crystal causes opposite strains for each of these domains via piezoelectric effect. It was shown that ferroelectric crystals with specifically tailored domain structures might have quite different and even new piezoelectric properties in comparison with monodomain ferroelectrics /3/,/4/.

The tensor of nonlinear optical coefficients is also of the third rank and its symmetry coincides with the tensor of piezoelectric coefficients. The sign of the nonlinear coefficient **d** of macroscopically polarized regions depends on the spontaneous polarization direction and it is opposite for ferroelectric 180°-domains. This enables to realize the idea of QPM proposed by Bloembergen /2/ by introducing into ferroelectric crystal bulk a specific domain configuration where the spontaneous polarization changes its direction alternatively. Several ideas were proposed to find another domain configurations for optical frequency tuning using ferroelectric domain engineering. Multi-grating domain design implemented in the works /5/,/6/ included fabrication of 25 domain grating sections ranged the domain grating period from 26 to 32 mm in 0.25 μm increments on one $LiNbO_3$ plate. This optical chip allowed generating OPO output tuned across the entire mid-IR transparency of $LiNbO_3$ from 1.98-1.36 mm in the signal branch and in the range 2.30-4.83 μm in the idler branch. In other work /7/ the continuous tuning was reached using fan-out grating design. Another approach where Fibonacci-based structures provided by aperiodic domain configurations /8/ were considered for simultaneously phase matching two interactions.

So far, ferroelectric domains, instead of deteriorating figures of merit, which occurs for instance in pyroelectric or electrooptic devices, demonstrate a unique ability to observe new and useful piezoelectric and optical properties based on specifically fabricated domain configurations. It paved the way for development of the new field-ferroelectric domain engineering /1/.

3. Domain broadening in ferroelectric domain structures

The most promising results in fabrication of the engineered domain structures were achieved by electrical poling technique, when pulsed electric switching field is applied to a monodomain ferroelectric crystal through a photolithographically patterned electrode /1/. The fabricated domain structure represents strip-like domains of a micrometer-scale width with straight, vertical domain boundary walls throughout the crystals bulk. However, despite of a strictly defined electrode pattern, the inverted domains are often wider than the deposited electrode strips. This unwanted domain broadening effect makes it especially difficult to tailor dense domain patterns with small periods. The conversion efficiency of SHG and OPO strongly depends on quality of the tailored domain grating structure. Various types of deviations from the perfect geometrical periodici-

ty and changes of the duty cycle and their influence on the conversion efficiency of SHG were theoretically analyzed /9/.

The domain broadening is induced by propagation of the growing reversed domain to the region, which is beyond the region limited by the switching patterned electrodes. It may be caused by widening of the growing domains due to a high tangential field between electrode strips, by bulk domain nucleation or by anisotropy of the domain walls velocity. Tangential domain widening in KTP crystals has been studied in the work /10/. Two other mechanisms of domain broadening such as anisotropic domain walls propagation and bulk nucleation of ferroelectric domains are considered.

3.1 Anisotropy of domain wall velocity

The shapes of the domain strips in the domain-grated structures induced by applied electric field depend on shapes of domain nuclei and anisotropy of the domain wall velocity. It is clear, that both factors may cause a distortion of the fabricated domain structures especially for the dense domain gratings. Our experiments have shown that KTP and isomorphic crystals demonstrate extremely high anisotropic growth of the inverted ferroelectric domains /11,12/. Dynamic ferroelectric domain shapes of the KTP crystals were studied by partial polarization reversal of a sample with uniform silver paint solid contacts applied to both polar Z-faces. The reversed domains are strongly elongated in the Y-axis direction (Figure 1). This experiment confirms that orientation of the patterned electrode along the principal axis of the ferroelectric crystals plays a crucial role in tailoring the domain configurations in these crystals. In the case of orientation of the electrode strips along the Y-axis the perfect domain grating can be fabricated (Figure 2(a)). When the electrode strips are oriented along the X-axis, the reversed domain structure does not correspond to the switching electrode pattern (Figure 2(b)). The reversed domains grow in the direction of the Y-axis and coalesce under the dielectric layer. It leads to complete polarization reversal of the ferroelectric sample.
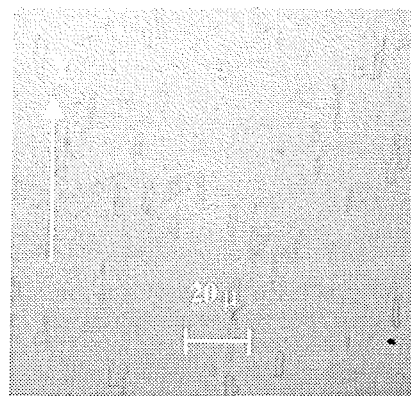

Figure 1: *The optical microphotography of the etched Z-plane of KTP crystal after partial polarization switching*
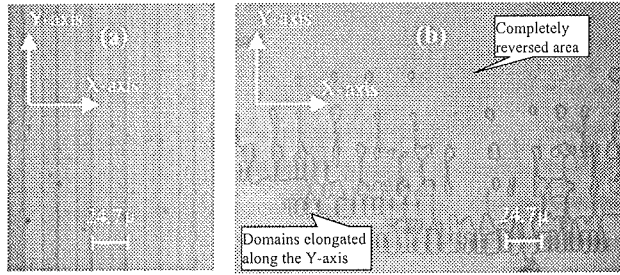
Figure 2: *The effect of the orientation of the patterned switching electrode on fabricated periodically poled structure: (a) - domain pattern obtained with the patterned electrode strips parallel to the Y-axis, (b) - domain pattern obtained with the patterned electrode strips parallel to the x-axis.*

The observed anisotropy of the domain wall velocity may be explained by a classic approach applied to the polarization reversal effect by Miller and Weinreich [/13/. They considered the mechanism of the sidewise motion of domain walls in weak fields as nucleation and subsequent growth of triangular reversed steps on 180^0 walls. The equation obtained by Miller and Weinreich for the domain walls velocity /13/ may be written in the form

$$v = v_\infty \exp(-\alpha b^{3/2})$$ (1)

where *b* is the lattice constant and *a* is a constant depending on the parameters of the two-dimension nuclei, the applied field density and the temperature. The lattice constants in tetragonal $BaTiO_3$ are equal along the X and Y directions. As a result, the 180^0 domains observed by Miller in $BaTiO_3$ possessed rectangular forms /14/. In contrast, the crystal structure of the KTP crystals is extremely anisotropic. The lattice constant along X-axis is 12.814 Å, and it is twice less in the Y-direction: 6.404 Å /18/. According to the expression (1), reversed domains in KTP crystals should grow in the Y-axis direction, where the lattice constant is minimal. This leads to the anisotropic domain form in these crystals observed in our experiments (Figures 1, 2).
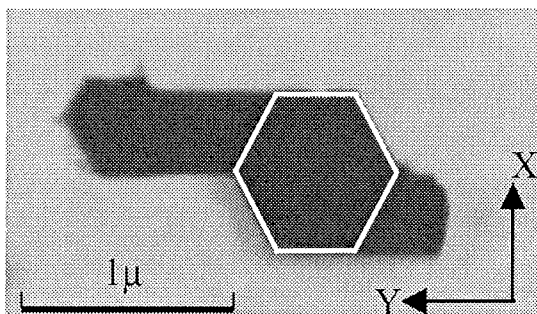

Figure 4: *Atomic Force Microscopy image of a reversed KTP micro-domain.*

Highly pronounced anisotropy of inverted domains makes KTP and its isomorphs especially attractive for fabrication of fine domain structures with periods of 1-2 micron. Our experiments using Atomic Force Microscopy demonstrated that domain nuclei of a sub-micron size in KTP crystals have a hexagonal form (Figure 3). The sidewise expansion of the reversed domains occurs by anisotropic motion of the hexagon sides in the Y-axis direction (Figure 3). Due to this extremely anisotropic motion of domain walls of sub-micron nuclei, domains with micron width oriented in the Y-axis direction could be tailored. The quasiperiodic structure for multiple nonlinear interactions with a minimal domain width 2 micron was successfully fabricated and showed good optical results. Hexagonal-shaped microdomains were observed by Dougherty *et al.* in $Pb_5Ge_3O_{11}$ ferroelectric crystals /15/.

3.2 Bulk domain nucleation

Another contribution to the domain widening is a bulk domain nucleation. According to the existing concepts /16/, compensation of the depolarization energy is a crucial condition for formation of domain nuclei with reversed spontaneous polarization. The depolarizing field may be screened by charges brought through the external circuit via switching electrodes or by the free charges existing in the crystal bulk.

Conventionally, most of ferroelectric materials are considered as ideal dielectrics with a very large dielectric relaxation time $\tau = \varepsilon\varepsilon_0/\sigma$ (e is the dielectric permittivity, and σ is the dc conductivity). TGS crystals ($\sigma \sim 10^{-18} \Omega^{-1}cm^{-1}$, $\tau \sim 10^6$ sec) provide a good example. In this case the polarization switching time τ_{sw} which may be several milliseconds, is much shorter than the time constant of the internal screening $\tau(\tau_{sw} \ll \tau)$ and only the external compensation via switching electrodes process occurs. The dielectric film inserted between the switching electrode and a TGS sample totally prevents the external screening of the depolarizing field and as a result polarization switching /17/.

KTP crystals possess a very high room-temperature ionic conductivity as compared to classic ferroelectrics, due to the structure peculiarities of these crystals /18/. This conductivity is ascribed to hopping of highly mobile potassium ions through the channels existing along the polar axis. Our study of dielectric properties of these crystals shows /19/, that the cation mobility reduces with decrease of the temperature, and at the definite temperature region around $T = 170$ K the ionic conductivity is frozen. Measurement of the dc conductivity values of KTP crystals showed $\sigma = 5 \times 10^{-13}$ $\Omega^{-1}cm^{-1}$ at the low temperature ($T = 170$ K) and $\sigma = 5 \times 10^{-6}$ $\Omega^{-1}cm^{-1}$ at room temperature ($T = 300$ K).

Our experiment of polarization switching of KTP crystals with a single strip of Ti electrode (5-µm width) deposited on the +Z face of the sample confirmed that the bulk domain nucleation in these crystals is caused by their high ionic conductivity. The −Z face of the samples was coated uniformly by a silver paint electrode. The polarization switch-
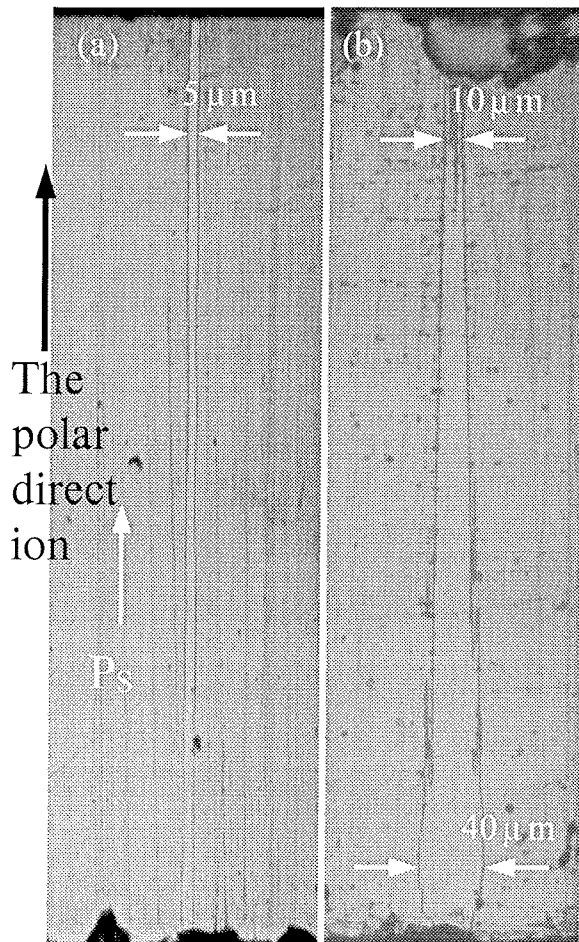
Figure 5: *The view of the KTP sample polished at 45°
to the z-plane and selectively etched after
polarization reversal (a) at T = 170 K,
(b) at T = 300 K.*

ing was performed by application of square pulses of high voltage with amplitude exceeding the coercive field, where the switching time of KTP crystals is 0.1-10 ms /11,20/. At T = 170 K $\tau_{sw} \ll \tau = \varepsilon\varepsilon_0/\sigma \sim 1$ s. At this temperature, as could be seen from Figure 4(a), bulk nucleation does not take place. The domain propagates via the crystal bulk without any broadening preserving its initial width of 5 µm. The sample switched at room temperature ($\tau_{sw} \gg \tau \sim 10^{-5}$ s) demonstrates a strong widening of the reversed domain (Figure 4(b)). In the lower part of this sample the width of the reversed domain is 40 µm, which exceeds by eight times the width of the electrode strip. In this case mobile K^+ cations contribute effectively to the internal screening of the depolarization field. Small nuclei, which were observed in the crystal bulk near the walls of the reversed domain, confirm the bulk nucleation origin of the domain broadening.

4.   Conclusion

Fabrication of engineered domain structures allows observing new physical properties of ferroelectric crystals. The physical origin of this phenomenon is changes in a macroscopic symmetry of the domain-patterned ferroelectrics. It is shown that domains growing in KTP crystals in low electric field are highly anisotropic: they are elongated along the Y-axis direction. This could be explained by the difference in lattice constants along the X and Y principal axes. The shape of the micro-domains of KTP possesses a hexagonal form. A strong influence of the ionic conductivity on the polarization reversal in KTP crystals was observed. KTP crystals reversed at room temperature demonstrate strong domain broadening as a consequence of bulk domain nucleation.

5.   References

/1./   G. Rosenman, A. Arie and A. Skliar, *Ferroelectrics Review*, **1**, 4 (1998).

/2./   J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Perhsan, *Phys. Rev.*, **127**, 1918 (1962).

/3./   R. E. Newnham and L.E. Cross, *Ferroelectrics*, 10, 269 (1976)

/4./   R. E. Newnham, C. S. Miller, L.E. Cross and T. W. Cline, *Phys. Stat. Sol. (a)*, **32**, 69 (1975)

/5./   L. M. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, W. R. Bosenberg, *SPIE*, **2700**, 216 (1997)

/6./   L. M. Myers, R. C. Eckardt, M. M. Fejer, R. L. Byer, W. R. Bosenberg, *Opt. Lett.*, **21**, 591 (1996)

/7./   P. E. Powers, T. J. Kulp, and S. E. Bisson, *Opt. Lett.*, **23**, 159, (1998)

/8./   K. Fradkin-Kashi, A. Arie, *IEEE Quant Electron*, **35**, (1649) 1999

/9./   M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, *IEEE J. Quantum Electron.*, **28**, 2631 (1992)

/10./   G. Rosenman, A. Skliar, D. Eger, M. Oron, M. Katz, *Appl. Phys. Lett.* **73**, 3650 (1998).

/11./   P. Urenski, M. Lesnykh, Y. Rosenwaks, G. Rosenman, M. Molotskii, *J. Appl. Phys.* **90**, 1950 (2001).

/12./   P. Urenski, G. Rosenman, M. Molotskii, *J. Mater. Res.* **16**, 1493 (2001).

/13./   R. C. Miller and C. Weinreich, *Phys. Rev.*, **117**, 1460 (1960).

/14./   R. Miller, *Phys. Rev.* **111**, 736 (1958.)

/15./   J. Dougherty, E. Sawaguchi, L. Cross, *Appl. Phys. Lett.* **20**, 364 (1972).

/16./   R. Landauer, *J. Appl. Phys.* 28, 227 (1957).

/17./   V.D. Kugel, G. Rosenman, *J. Appl. Phys.* **80**, 5256 (1996).

/18./   L. K. Cheng and J. D. Bierlein, *Ferroelectrics* **142**, 209 (1993).

/19./   P. Urenski, N. Gorbatov, G. Rosenman, *J. Appl. Phys.* **89**, 1850 (2001).

/20./   G. Rosenman, A. Skliar, M. Oron, M. Katz, *J. Phys. D.* **30**, 277 (1997).

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