Pyroelectric field-assisted domain engineering in lithium niobate and lithium tantalate using femtosecond laser pulses

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Introduction

Engineered domain structures play an essential role in nonlinear optics for quasi-phase-matched parametric processes [1]. There is a great interest in domain inversion techniques based on all-optical processes which do not require external electric fields [2]. Lately, promising results have been obtained in structuring the nonlinearity in lithium niobate ($LiNbO_3$) with focused femtosecond laser pulses. These include all-optical poling [3] and $\chi^{(2)}$ depletion [4, 5]. However, these methods only allow for small feature-sized domain inversion or show increased absorption with reduced efficiency. A promising approach to overcome the aforementioned limitations is based on our recently discovered pyroelectric field-assisted domain inversion process that can be used to switch ferroelectric domains in the volume of magnesium doped $LiNbO_3$ without using an external applied electric field [6]. The process works as follows (see Fig. 1). First, focused femtosecond laser pulses induce permanent defects along the polar axis. Then, the crystal is heated up above 200 °C. During cooling, the domain inversion is driven by a space-charge field that locally exceeds the threshold field of domain nucleation. After this heating-cooling cycle, ferroelectric domains are inverted below and above the defects that act as seeds. Domain inversion occurs if a certain pulse energy and defect length are exceeded. However, there is no detailed study on the role of temperature and domain spacing in this process. The latter one is essential for the fabrication of two-dimensional nonlinear photonic structures because a certain length of a reciprocal lattice vector is required for a specific quasi-phase-matching process.

Here, we examine the pyroelectric field-assisted domain inversion in $LiNbO_3$:Mg, dependent on the temperature for 2D rectangular lattices of different periods. We measure the fraction of domains that are inverted and the domain diameters.

Pyroelectric field-assisted domain inversion

We fabricate two-dimensional lattices of ferroelectric domains by patterning magnesium-doped $LiNbO_3$ and near-stoichiometric lithium tantalate crystals with femtosecond laser pulses ($\lambda = 1030\,\mathrm{nm}$) with a pulse width of 250 fs, a repetition rate of 1 kHz and energies of up to 400 nJ. We create on different samples 2D nonlinear photonic structures with periods of (6 µm, 10 µm, 15 µm) x 6.3 µm. The crystals are then heated to various temperatures in the range of 50 °C to 400 °C and cooled down. We investigate the effect of temperature and seed spacing on the number and size of inverted domains in $LiNbO_3$:Mg. Čerenkov second-harmonic generation microscopy allows visualizing the generated ferroelectric domains and laser-induced seeds in 3D [7]. Measurements with different electrical terminations

of the crystal surfaces reveal the influence of surface charges during the domain formation process.

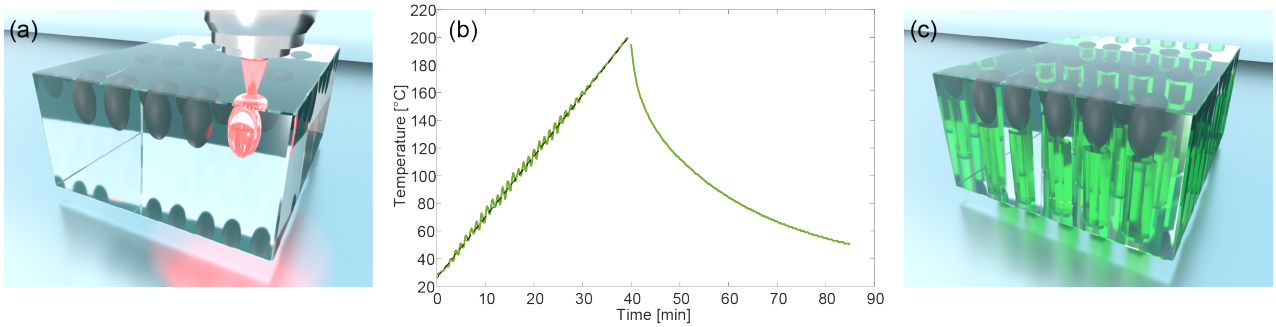

Figure 1: Scheme of the experimental procedure. (a) Permanent defects (grey) are induced with focused femtosecond laser pulses (red) along the optical axis. (b) The sample is heated to elevated temperatures and cooled down to room temperature. (c) After cooling down, ferroelectric domains (green) are inverted below and above the defects.

Conclusions

We have identified a threshold temperature and determined an optimal temperature regime between 220 °C and 300 °C in $LiNbO_3$:Mg. In this temperature range, all domains can be inverted in a 2D rectangular lattice with periods of 15 μm×6.3 μm. Smaller lattice periods and lower temperatures result in fewer inverted domains. Above 300 °C the already inverted domains switch back in the center of the lattices. The average diameter of the central domains is approximately 2.4 μm. This size is almost independent of temperature and lattice periods. The electrical termination of the crystal surfaces has no significant influence on the domain formation process unless the crystal is completely short-circuited. In this case, no domains are inverted, indicating that surface charges are essential for the inversion process.

References

[1] A. Arie and N. Voloch, Laser Photonics Rev. **4**, 355–373 (2010).
[2] C. Ying, A. Muir, C. Valdivia, H. Steigerwald, C. Sones, R. Eason, E. Soergel, and S. Mailis, Laser Photonics Rev. **6**, 526–548 (2012).
[3] X. Chen, P. Karpinski, V. Shvedov, K. Koynov, B. Wang, J. Trull, C. Cojocaru, W. Krolikowski, and Y. Sheng, Appl. Phys. Lett. **107**, 141102 (2015).
[4] D. Wei, C. Wang, H. Wang, X. Hu, D. Wei, X. Fang, Y. Zhang, D. Wu, Y. Hu, J. Li, S. Zhu, and M. Xiao, Nat. Photonics **12**, 596–600 (2018).
[5] J. Imbrock, L. Wesemann, S. Kroesen, M. Ayoub, and C. Denz, Optica **7**, 28 (2020).
[6] J. Imbrock, H. Hanafi, M. Ayoub, and C. Denz, Appl. Phys. Lett. **113**, 252901 (2018).
[7] Y. Sheng, A. Best, H.-J. Butt, W. Krolikowski, A. Arie, and K. Koynov, Opt. Express **18**, 16539 (2010).