Latent ultrafast laser-assisted domain inversion in congruent lithium niobate

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The combination of light with external electric fields has been successfully used for the domain engineering of ferroelectric lithium niobate crystals [1,2]. It has been shown that *simultaneous* illumination and application of an external electric field can achieve, e.g. in the case of MgO-doped lithium crystals, coercive field reduction up to 98% using fs-pulsed light [2].

In the case of undoped congruent lithium niobate, however, it has been observed and it is reported here that *photo-induced coercive field reduction also occurs in a latent manner* whereby the application of the electric field is delayed with respect to the illumination of the crystal. Furthermore, the local coercive field reduction becomes fixed after the first poling cycle. Hence, the initially illuminated and domain inverted regions will re-invert at lower voltages for subsequent poling cycles. The most significant implication of the latency is the decoupling of the laser illumination and E-field application steps which significantly simplifies the experimental setup and allows for high resolution light patterning, e.g. using a phase mask.

The second harmonic of the ultra-fast laser system (Coherent: Mira oscillator, RegA amplifier, repetition rate of 250 kHz, pulse duration ~130 fs) at $\lambda = 400$ nm was focused onto the –z face of the crystal to a spot size of approximately 45 μ m. The average power of the illuminating laser beam was 10 mW, 20 mW and 40 mW which correspond to a peak intensity of 4.5 GW/cm², 9 GW/cm² and 18 GW/cm² respectively. Congruent lithium niobate samples of 500 μ m thickness were initially "conditioned" by a sequence of five "dark" poling cycles before illumination to ensure consistency of the forward and reverse values of the intrinsic coercive field. In this investigation a transparent cell was used for the illumination and poling of the crystals which allowed *in situ* observation of the domain formation (via stress-induced birefringence and electro-optic refractive index contrast at the inverted domain boundaries).

The latent laser-assisted inverted ferroelectric domains, shown in Figure 1a, have all been poled at the same voltage. They correspond however to different time delays between the illumination time and the voltage application time, with the far right being the most recent. It is obvious from this figure that the domain spreading is a function of the illumination-voltage time delay. Figure 1b plots the results of a more comprehensive investigation of the resulting domain area (measured after brief etching of the crystal in HF acid) as a function of the time delay before the application of voltage. Each spot was illuminated for 30s, with a 30 s delay between subsequent illuminations. 30 s after the last exposure, the voltage was ramped at a rate of \sim 130 V/s, maintained at the maximum voltage V = 4 kV for 300 s and finally ramped down to 0 kV at a rate of \sim 130 V/s.

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A full study of the latent coercive field modification and domain expansion dynamics will be presented.



Figure1: (a) Ferroelectric domains of different areas corresponding to different time delays between illumination and voltage application, the far right being the most recent. (b) Ferroelectric domain area vs. voltage application time delay for three different illuminating powers with an applied voltage of 4 kV.

References.

[1] M. Fujimura, T. Sohmura, and T. Suhara, "Fabrication of domain-inverted gratings in MgO:LiNbO₃ by applying voltage under ultraviolet irradiation through photomask at room temperature," Electron. Lett., **39**, 719-21 (2003). [2] C.E. Valdivia, C.L. Sones, S. Mailis, J.D. Mills, and R.W. Eason, "Ultrashort-pulse optically-assisted domain engineering in lithium niobate," Ferroelectrics, 340, 75-82 (2006).