

## Ultrafast ferroelectric switching: what are the limitations?

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Manipulation of the atomic lattice using high power single-period THz pulses has been proven already directly by time-resolved X-Ray diffraction (XRD) techniques in different types of dielectrics [1-2]. Possibility to contactless displacement of polar ion is particularly important in ferroelectrics, where the frequency limitation arises due to the conventional circuit limitations for coupling electrical pulses through electrode structures on a sub-100-picosecond (ps) time scale. At the same time, the dynamics of electric polarization lies at the heart of ferroelectric-based ultrafast next-generation piezoelectric, electro-optic, and nonvolatile memory devices. Along with XRD, optical second harmonic generation (SHG) is widely used for justification ferroelectric switching under THz pulse [3-4].

In ferroelectrics, analogously to magnetic materials, short electromagnetic pulses may cause coherent collective excitations. In ferroelectric these are phonons, including the soft mode ones. Amplitude of such excitations is much larger than in case of thermal excitations. Excitations take place coherently over macroscopic area of illuminating spot (from tens to thousands of square microns). In this case, one can consider dynamical switching of ferroelectric polarization within the electromagnetic pulse duration following by relaxation. If the amplitude of coherent oscillations is high enough to transfer the process into highly nonlinear regime, than oscillating polar ion may relax to the different position of two-minima potential regarding to its initial position. In contrast to coherency of oscillation, these relaxations are stochastic. Thus, polarization switching, if takes place, is temporally limited: in dynamical range – to the reverse soft mode frequency; for polarization reversal – to the pulse duration.

In this presentation, we focused on theoretical justification on the conditions (parameters of ferroelectric crystal and electromagnetic pulse) providing both regimes of polarization switching within the Landay-Khalatnikov (LK) model. The model describes not only the ion displacements, but also the nonlinear-optical response of the materials. We took real parameters of the potential function of different crystals and considered LK equations with 1 ps THz pulse as external force. In this way, SrTiO<sub>3</sub>, KTaO<sub>3</sub>, BaTiO<sub>3</sub>, (BaSr)TiO<sub>3</sub> were investigated. We make search for parameters and materials which can provide conditions for both types of switching. In description of the non-linear optical response we take into account the initial non-polarized multidomain state. Due to the nonidentical initial conditions for such a state, the solutions of the Landau–Khalatnikov equation are also nonidentical, and their interference gives a nonzero second harmonic field under THz electric field. We consider the temperature dependences of the second harmonic in the framework of such a model.

Experimentally, we performed a comparative study of THz field induced SHG in different ferroelectrics, incipient ferroelectric and multiferroic materials: SrTiO<sub>3</sub>, KTaO<sub>3</sub>, (BaSr)TiO<sub>3</sub>, BiFeO<sub>3</sub>, (BaSr)TiO<sub>3</sub>/BiFeO<sub>3</sub>. In these materials using time-domain spectroscopy (THz pump–optical probe), studies have been carried out of the temporal dependences of the intensity of the optical second harmonic. The highest switching efficiency was revealed in (BaSr)TiO<sub>3</sub> multidomain films near the phase transition where the soft mode is very wide and overlaps with the THz pulse in a frequency domain. In this material, we have proven the dynamic switching of the ferroelectric polarization with a THz pulse starting from a field of 300 MV/cm.

1. F. Chen, Y. Zhu, et al, *Phys. Rev. B* **94**, 180104(R) (2016).
2. C. Hauf, A.A.H. Salvador, et al, *Structural Dynamics* **5**, 024501 (2018)
3. E. Mishina, K. Grishunin, A. Sigov, et al, *Ferroelectrics* **532**, 199 (2018).
4. K. Grishunin, V. Bilyk, et al, *Scientific Reports* **9**, 697 (2019).