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► **To cite this version:**

Vasily V Temnov, Paolo Vavassori. All-optical polarization switching in ferroelectrics. *Nature Photonics*, 2024, 18, pp.529 - 530. 10.1038/s41566-024-01452-9 . hal-04789851

HAL Id: hal-04789851

<https://hal.science/hal-04789851v1>


Submitted on 19 Nov 2024

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All-optical polarization switching in ferroelectrics

Vasily V. Temnov & Paolo Vavassori

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Ultrafast optical experiments using narrow-bandwidth tunable IR laser pulses enable nonvolatile all-optical switching of ferroelectric polarization in BaTiO₃ in the epsilon-near-zero regime.

Given the relevance of boosting the speed of data recording, investigations into the pathways of nonvolatile all-optical switching of order parameters in solid state systems using single ultrashort laser pulses have remained topical for decades^{1,2}. The fundamental problem lies in the fact that the transition to the final state is governed by various non-equilibrium dynamics on ultrafast, femtosecond-to-microsecond time scales. In ferromagnetic systems, fine-tuning the laser-material interactions through small magneto-optical effects allows one to elucidate the dominant switching mechanism³.

In the specific case of ferroelectric materials, ferroelectric domains are linked to distinct atomic configurations⁴, suggesting the necessity of ultrafast control over atomic configurations within a unit cell. An early theoretical study suggested that the use of properly shaped electric fields of THz pulses could be used to move ions in ferroelectric crystals from their positions in an initial domain orientation along well-defined collective microscopic paths into the positions they occupy in a new domain orientation⁵. More recently, the excitation of large-amplitude IR-active phonons in lithium niobate (LBO) was sufficient to create transient atomic configurations close to the switching regime.

Although nonvolatile ferroelectric switching was not able to be demonstrated in this experiment, the work clearly highlighted the importance of nonlinear optical interactions mediated by optical phonons in experiments of that kind. Permanent and reconfigurable polarization switching in LBO has been achieved in a scanning femtosecond laser writing experiment using a MHz-repetition rate laser source, whereas the switching was largely affected by thermoelectric fields caused by temperature gradients along the direction of laser scan⁶. Until now it was not known which of these distinct mechanisms is pivotal to achieve the nonvolatile ferroelectric switching on demand using single ultrashort laser pulses.

Now, writing in *Nature Photonics*, Kwaaitaal and co-workers investigate the dynamics of ferroelectric polarization in a perovskite-type ferroelectric material BaTiO₃ irradiated by bursts of ultrashort IR pulses delivered by a free electron laser (FELIX facility in Nijmegen)⁷. The experiment represents a quasi-single-shot regime, as each position of the sample is irradiated by 8- μ s-long bunches of mid-IR narrow-bandwidth radiation consisting of 200 picosecond pulses separated by 40 ns time intervals. The wide tunability of the free electron laser wavelength range between 7 and 28 μ m, together with the high pulse energies in the μ J range, is an ideal condition in which to study the

dynamics of laser-driven ferroelectric polarization across two optical phonon resonances at 14 and 21 μ m, respectively.

In the study, special care has been taken with regard to the development of different experimental methods necessary to clearly and unambiguously identify ferroelectric domains. Whereas polarization microscopy was sufficient to identify the 90° domains (Fig. 1b), the second harmonic generation microscopy was necessary to visualize the 180° domains (Fig. 1c).

Quantitative analysis of such microscopy images made possible the measurement of the switched area as a function of excitation wavelength (Fig. 1d). Notably, the authors have been able to correlate the maxima of the switching efficiency with the zeros of the dielectric function – that is, with the realization of epsilon-near-zero (ENZ) conditions naturally emerging from the peculiar phononic properties of BaTiO₃. As a result, the authors have shown, on the one hand, that BaTiO₃ implements what they define as a natural “phononic” ENZ material⁸, and, on the other hand, that the strongly amplified light–matter interaction at the ENZ conditions gives rise to the long-sought and so far elusive non-volatile all-optical switching of its ferroelectric polarization.

The exact microscopic switching mechanism remains largely unclear. Preliminary time-resolved studies appear to indicate ultrafast reorientation of ferroelectric domains. After having applied several time-resolved techniques with instrumentation-limited time resolution, the authors estimate the upper limit of the switching time to be around 3 ns.

In order to explore the photonic aspects of the switching phenomenon further, a better understanding of the unique and exclusive optical properties of ENZ materials is needed. Figure 1f shows strong dispersion effects across both resonances, with a remarkable slowing down of light quantified by the up to fortyfold increase in the group index for the excitation radiation. In such a slow-light regime, the interaction time between the mid-IR pulses and ferroelectrics can greatly exceed the pulse duration in vacuum. This remarkable property may indeed pave the way for an understanding of the mechanisms leading to the switching phenomenon through the prism of coherent control⁵ or nonlinear optical^{8,9} models. Beyond these readily established theoretical concepts, the authors developed a simple theoretical model explaining how laser heating of the sample produces the quasi-static Gaussian distribution of thermal strain, which in turn generates the polarization profiles matching those of the switched domains. With all these exciting observations the compelling quest towards the ultimate speed limit of ferroelectric switching remains open. Given the fact that table-top low-repetition-rate μ J-energy mid-IR OPA-based femtosecond laser sources are readily available¹⁰, the search for exact pathways of ultrafast ferroelectric switching in the true single-shot regime can be tackled by optical femtosecond time-resolved polarization sensitive microscopy techniques¹¹.

The strong coupling between lattice, charge, spin, and orbital degrees of freedom offers new vistas and a broad playground for a variety of exotic and technologically relevant properties that might be

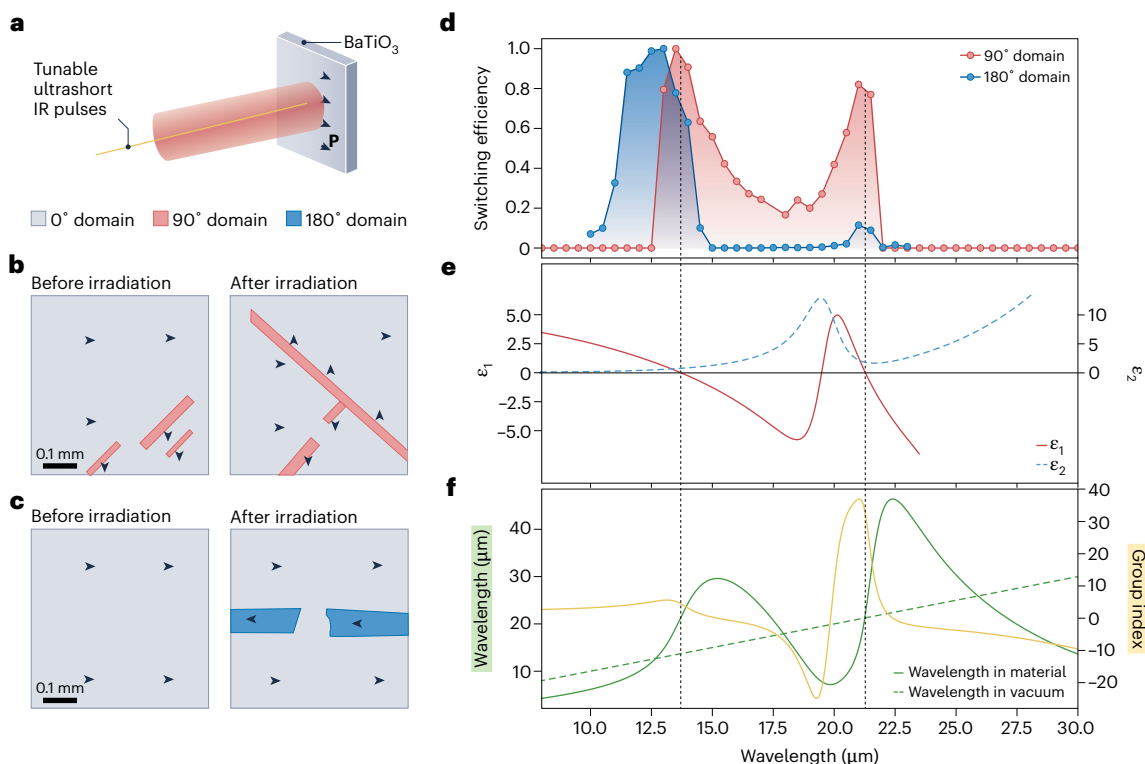


Fig. 1 | Experimental demonstration of ultrafast IR-laser driven ferroelectric switching in BaTiO₃. **a–c**, Sample illumination with femtosecond IR laser pulses results in a permanent reorientation of 0° polarization domains into 90° domains (**b**) and 180° domains (**c**). **d**, The normalized area of switched domains depends on the excitation wavelength. **e**, The maxima of switching correlate with the

epsilon-near-zero (ENZ) condition as quantified by the spectral dependence of the linear dielectric function with two resonances around 13 μm and 21 μm. **f**, The ENZ regime is accompanied by strong dispersive effects, most pronounced in the up to fortyfold increase in the group index (slow light) at the second resonance. Figure adapted from ref. 7, Springer Nature Ltd.

tuned in the same way. The general concept itself – namely, the occurrence of material-inherent ENZ conditions to enable ultrafast all-optical switching of order parameters – is potentially disruptive and capable of reaching far beyond the specific case of a ferroelectric illustrated here, provided one can find ENZ materials in which the property enabling the occurrence of ENZ conditions is directly or indirectly coupled to the order parameter to be tuned.

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Published online: 6 June 2024

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Competing interests

The authors declare no competing interests.