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RESEARCH ARTICLE

Modeling of resonant excitation of ferroelectric lattice subsystem by terahertz radiation under nonequilibrium conditions

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Abstract

Objectives. The search for a fundamentally new, fast, and least dissipative method for controlling the ferroic order parameter is a pressing and ambitious task of basic and applied research on the development of low-dissipation and high-speed functional elements of information systems operating at terahertz (THz) frequencies for 6G[1] network technologies. The aim of the work is to study the conditions for modulating ferroelectric polarization using a short THz pulse. This will also include the influence of additional factors on the efficiency of the THz-induced dynamics of the ferroic order parameter, such as stationary heating and the application of an additional electric field to the ferroelectric.

Methods. The numerical simulation of resonant excitation of the lattice subsystem by THz radiation was performed for a $Ba_{0.8}Sr_{0.2}TiO_3$ ferroelectric film using the Landau–Khalatnikov equation system, and the equation of phonon mode oscillations with the phonon–phonon interaction as a driving force. The novelty of the proposed approach lies in the interaction of the THz pulse with a previously coherently excited phonon mode with a significant amplitude.

Results. The time dependencies were presented for the polarization and the effective amplitude of infrared-active mode oscillations with varying THz field parameters. The results also included constants of expansion of the thermodynamic potential of the nonequilibrium state in powers of order parameter in various exposure modes, including temperature changes and the application of an additional external electric field.

Conclusions. The approach proposed herein describes polarization switching under the action of a THz pulse with preliminary excitation of a coherent phonon by a femtosecond optical pulse. The most important parameter when determining the threshold effect of a THz pulse on phonon excitation is pulse energy (amplitude). In the region of small amplitudes, an increase in temperature does not exacerbate switching conditions.

Keywords: terahertz radiation, ferroelectrics, polarization, resonant excitation, phonon modes

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[1] Sixth generation of mobile communications technology.

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НАУЧНАЯ СТАТЬЯ

Моделирование резонансного возбуждения терагерцевым излучением решеточной подсистемы сегнетоэлектрика в условиях неравновесного состояния

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Резюме

Цели. Поиск принципиально нового наиболее быстрого и наименее диссипативного способа управления ферроидным параметром порядка является актуальной и весьма амбициозной задачей фундаментальных и прикладных исследований в области разработки малодиссипативных и быстродействующих функциональных элементов информационных систем, оперирующих на терагерцевых (ТГц) частотах, для сетевых технологий 6G[2]. Целью работы является исследование условий модуляции поляризации сегнетоэлектрика при помощи короткого ТГц-импульса, в т.ч. изучение влияния дополнительных факторов на эффективность ТГц-индуцированной динамики ферроидного параметра порядка.

Методы. Численное моделирование резонансного возбуждения ТГц-излучением решеточной подсистемы проводилось для сегнетоэлектрической пленки титаната бария-стронция $Ba_{0.8}Sr_{0.2}TiO_3$ на основе системы уравнений, состоящей из уравнения Ландау – Халатникова и уравнения колебаний фононной моды, где в качестве вынуждающей силы выступает фонон-фононное взаимодействие. Новизна подхода заключается во взаимодействии ТГц-импульса с ранее когерентно возбужденной фононной модой, имеющей значительную амплитуду.

Результаты. Представлены временные зависимости поляризации, а также эффективной амплитуды колебаний инфракрасно-активной моды при варьировании параметров ТГц-поля и констант разложения термодинамического потенциала неравновесного состояния в виде ряда по степеням параметра порядка в разных режимах воздействия, включая изменение температуры и дополнительное воздействие внешнего электрического поля.

Выводы. Предложенный подход описывает переключение поляризации под действием ТГц-импульса при предварительном возбуждении когерентного фонона фемтосекундным оптическим импульсом. Наиболее важным параметром, определяющим пороговое воздействие ТГц-импульса на возбуждение фонона, является энергия (амплитуда) импульса. В области малых амплитуд увеличение температуры не приводит к ухудшению условий переключения.

Ключевые слова: терагерцевое излучение, сегнетоэлектрики, поляризация, резонансное возбуждение, фононные моды

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[2] Sixth generation – шестое поколение мобильной связи. [Sixth generation of mobile communications technology].

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INTRODUCTION

The development of terahertz (THz) devices is one of the most pressing areas in modern micro- and nanoelectronics and photonics. In addition to the methods of THz spectroscopy and visualization for diagnostics of materials already widely used, THz-range systems and functional devices designed to monitor and control THz signals are currently attracting increasing attention [1, 2]. The use of the THz range (0.1–10 THz) for data transmission has been shown to provide high throughput and data density, and is considered to be the most promising approach for 6G[3] network technologies [3, 4].

One of the key challenges in this area is the search for new materials to ensure efficient optical and/or electrical control of the generation, detection, and propagation of THz radiation. Ferroelectric materials play an important role in the study of functional devices operating in the THz range. This is due to their response time, low dielectric loss, and tunability. As demonstrated in a number of studies, the ability to manipulate optically the permittivity of thin ferroelectric films allows for significant modulation depth in the THz range. This is of great importance for the development of all-optical and hybrid chips.

It is important therefore to excite the soft mode of a ferroelectric and use this excitation to switch the polarization of the ferroelectric by a short THz pulse. The first works on THz excitation by relatively low fields (to 100 kV/cm) reported on coherent excitation of small-amplitude polar modes in strontium titanate ($SrTiO_3$) crystals [5], relaxor ferroelectrics [6], and an organic ferroelectric [7]. The progress achieved in recent years in the development of THz sources capable of generating pulses (including single ones) of high power has made it possible to observe not only the excitation of coherent phonons, but also ultrafast (subpicosecond) polarization switching. Such switching was recorded either directly by X-ray diffraction methods [8–11] or by optical methods [12–14]. Recent studies have also discussed the effect of an ultrashort electromagnetic pulse leading to two types of polarization switching: permanent switching, such that the switching to the opposite polarization state under an external action persists after the end of the action; and dynamic switching, such that, after exposure to a THz pulse, the polarization state can switch to the opposite state and back several times. Then eventually the system relaxes to the initial state.

While the problem of the direct influence on the soft mode has been resolved in a certain sense, the influence of phonon–phonon interaction on the processes is still actively debated. A number of studies have proposed an approach based on the Landau–Khalatnikov equation [15]. In such studies the influence on the soft mode is considered within the framework of a model for coupling between the Raman and infrared-active (IR-active) modes [16, 17]. Mankowsky et al. [13] and Subedi [18] proposed describing ultrafast polarization switching processes using an approach based on a nonlinear coupling between the low-frequency, fully symmetric phonon mode A_1. This involves the motion of cations and anions of the material in such a way the electric polarization is changed, as well as the highest-frequency, and IR-active phonon mode. In this case, the switching processes are characterized in terms of the displacements of ions in the crystal lattice, described by the generalized coordinates Q_P and Q_IR. The coupling between the modes is proportional to the product of the generalized coordinate of the polar mode and the square of the generalized coordinate of the IR-active mode. This allows different switching conditions to be assumed for polarization vectors directed in opposite directions before the start of the action. However, Mertelj and Kabanov [19] indicated that such a representation is erroneousness due to the symmetry of the transformation. They proposed to consider the product of the squares of the generalized coordinates of the polar and IR-active modes as the main term. It was also noted that the products of the first powers of the generalized coordinates of the polar and IR-active modes also satisfy the symmetry conditions. However, when considered without the quadratic term, they do not explain the experimental dependencies presented by Mankowsky et al. [13].

The aforementioned models describing the modulation of polarization and its permanent switching yield unstable solutions. Small changes in any parameter cause an abrupt change in the properties of the system, as

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[3] Sixth generation of mobile communications technology.

described by the Landau–Khalatnikov equation, taking into account the strong nonlinearity of polarization near resonance [20]. Therefore, it is important to consider possible additional influences on the ferroelectric such as: heating, electric field, and optical illumination. A comprehensive solution to this problem will only be possible with the identification of physical mechanisms capable of achieving practical polarization switching using an electromagnetic pulse.

In this paper, we considered the processes of modulation of the polarization of a ferroelectric by a short THz pulse using barium strontium titanate $Ba_{0.8}Sr_{0.2}TiO_3$ (BST) as an example. An extension of the theoretical model [19] was presented using two terms in the expansion of the phonon–phonon interaction energy: the sum of the linear and quadratic terms over the product of the generalized coordinates of the polar and IR-active modes. The simulation took into account the conditions of experimental observation of the THz-induced modulation of the ferroelectric polarization in the optical pumping–THz probing geometry.

CALCULATION METHOD

Polarization switching under the action of a THz pulse with preliminary excitation of a coherent phonon by a femtosecond optical pulse

Let us consider a system consisting of unexcited ferroelectric cells with a two-minimum potential and an optically excited phonon in the field of a THz pulse. In this case, excitation of the soft mode via phonon–phonon interaction is energetically more favorable. Since the polarization of a ferroelectric is determined by the asymmetric displacement of cations and anions in the crystal structure, excitation of a coherent optical phonon increases the effective vibration amplitude of the corresponding group of atoms. This amplitude, which increases with increasing optical pulse intensity, will serve as the initial condition in the problem under consideration.

Let us also assume that a THz pulse, resonant with the ferroelectric's soft mode, is incident on the ferroelectric after some delay. Thus, the soft mode will be excited both directly by the THz pulse and through phonon–phonon interaction by an optical phonon. This in turn, was excited by femtosecond optical pumping. The question to be addressed is as follows: will this interaction be more effective than direct interaction with a single THz pulse, and will it lead to a higher probability of polarization switching in the ferroelectric?

Since BST is a multiaxial ferroelectric, its order parameter is multicomponent. In accordance with the approach published [13, 18, 19], we will restrict ourselves to considering the change in the polarization state under the action of the selective excitation of lattice vibrations by the electric field of a short THz pulse along the direction specified by the polarization plane of the THz radiation. In the initial state, we assume that the polarization caused by the total dipole moment of the set of cells considered in the region of action of the THz pulse is zero.

Taking into account Mertelj and Kabanov's comment [19], we can write the interaction energy of the polar and IR-active modes per unit volume in the following form:

$$W(\xi_P, \xi_{IR}) = j\xi_P\xi_{IR} + h\xi_P^2\xi_{IR}^2,$$

wherein ξ_P and ξ_{IR} are the effective coordinates of the polar and high-frequency phonon (IR-active) modes, respectively; and j and h are the expansion coefficients of $W(\xi_P, \xi_{IR})$ in powers of coordinates.

In accordance with Mertelj and Kabanov [19], since the coupling coefficient between the polar and IR-active modes depends significantly on the effective coordinate of the polar mode, the potential energy describing the phonon–phonon interaction must be physically significant, if the displacement of the polar ion relative to the equilibrium position is small. Within the framework of this model, the modulation of the effective coordinates does not exceed 5% of the lattice parameter. However, for the case of dynamic polarization switching, i.e., the hopping of a polar ion from one well to another, this restriction is imposed on the modulation of the effective coordinate ξ_P after such a hop.

Considering the definition of dielectric polarization $P = \frac{q_P\xi_P}{V}$ (where q_P is the effective charge, the displacement of which determines the excitation of the polar mode, and V is the cell volume), expressing the polar coordinate ξ_P through polarization, and introducing the notations $C_1 = j\frac{V}{q_P}$ and $C_2 = h\left(\frac{V}{q_P}\right)^2$, we can write the interaction energy in the following form:

$$W(P, \xi_{IR}) = C_1P\xi_{IR} + C_2P^2\xi_{IR}^2. \tag{1}$$

The equation of oscillations of the effective coordinate of the IR-active phonon mode is the equation of an anharmonic oscillator:

$$\ddot{\xi}_{IR} = 2\gamma_{IR}\dot{\xi}_{IR} + \omega_{IR}^2\xi_{IR} = -\frac{V}{m_{IR}}F_{IR}, \tag{2}$$

where γ_{IR} is the attenuation coefficient, and the right-hand side is determined by the derivative of free energy (1) with respect to the corresponding coordinate:

$$F_{IR} = -\frac{\partial W(\xi_P, \xi_{IR})}{\partial \xi_{IR}}.$$

In accordance with the Landau–Khalatnikov equation, the equation describing the dynamics of the order parameter under damping conditions takes the following form [20]:

$$\ddot{P} + \gamma\dot{P} + \frac{1}{\mu}(a_1 P + a_{11}P^3 + a_{111}P^5 + a_{1111}P^7) = \\ = \frac{1}{\mu}F_\text{P} + \frac{1}{\mu}E(t). \quad (3)$$

Here, $F_\text{P} = -\frac{\partial W(\xi_\text{P}, \xi_\text{IR})}{\partial \xi_\text{P}}$, $\mu = \frac{m_\text{P} V}{q_\text{P}^2}$, and $E(t)$ = $= E_\text{ext}(t) + E_1(t)$ is the electric field acting on the ferroelectric, equal to the sum of the external field $E_\text{ext}(t)$ and the depolarizing field $E_1(t)$.

The external field is a time-dependent electrical component of a picosecond THz pulse: $E_\text{ext}(t) = E_\text{THz}\mathrm{e}^{-\frac{4t^2}{\tau^2}}\frac{\sin z}{z}$, where $z = \omega_\text{THz}t$; and ω_THz and τ are the central frequency and the inverse attenuation of the THz field, respectively. These parameters were chosen to match the temporal shape of the calculated pulse with the experimental pulse, and are equal to $\omega_\text{THz} = 2\pi\cdot 1.5\ \frac{\text{rad}}{\text{s}}$ and τ = 0.5 ps. The depolarizing field is directly proportional to the polarization: $E_1(t) = E_\text{0,dep}\frac{P(t)}{P_\text{S}}$, wherein P_S is the spontaneous polarization corresponding to the position in one of the minima [21], and depolarizing field $E_\text{0,dep} = \frac{P_\text{S}}{\varepsilon\varepsilon_0}$.

Finally, using the introduced notations, we obtain a system of equations

$$\ddot{P} + \gamma_\text{P}\dot{P} + \frac{1}{\mu}(2a_1 P + 4a_{11}P^3 + 6a_{111}P^5 + 8a_{1111}P^7) = \\ = -\frac{1}{\mu}(C_1\xi_\text{IR} + C_2 P\xi_\text{IR}^2) + \frac{1}{\mu}E(t), \quad (4)$$

$$\ddot{\xi}_\text{IR} = 2\gamma_\text{IR}\dot{\xi}_\text{IR} + \omega_\text{IR}^2\xi_\text{IR} = -\frac{V}{m_\text{IR}}(C_1 P + C_2 P^2\xi_\text{IR}). \quad (5)$$

The experiment for which the simulation was performed can be carried out by means of the THz pumping–probing method in transmission geometry. Under THz pumping, several scenarios are possible (Fig. 1): (1) the THz field amplitude is insufficient for the effective charge to overcome the potential barrier, and the system oscillates around the initial minimum and returns to its initial state; (2) the effective charge overcomes the potential barrier but then returns to the initial minimum; and (3) the effective charge overcomes the potential barrier and remains at the opposite minimum of the double-well potential for a long time. The first two scenarios describe dynamic polarization switching, while the last describes a permanent one.

In the simulation using the system of equations (4)–(5), the time dependencies were calculated for the polarization and the effective amplitude of the IR-active mode oscillations parallel to the crystallographic axis *c*. The following parameters were varied: the THz field amplitude E_THz; the values of the phonon–phonon interaction constants C_1 and C_2; and the attenuation coefficient γ_IR of the IR-active mode.

The most important varying parameter was also the initial condition. This was determined by the intensity of optical radiation for the IR-active mode amplitude. The simulation used the parameters of the expansion of the thermodynamic potential in a series in powers of the order parameter (hereinafter referred to as the Landau expansion) for a $\text{Ba}_{0.8}\text{Sr}_{0.2}\text{TiO}_3$ film [22]: $a_1 = -0.13 \cdot 10^8$ J·m/C^2, $a_{11} = -1.33 \cdot 10^8$ J·m^5/C^4, $a_{111} = 1.03 \cdot 10^9$ J·m^9/C^6, and $a_{1111} = 3.09 \cdot 10^{10}$ J·m^{13}/C^8.

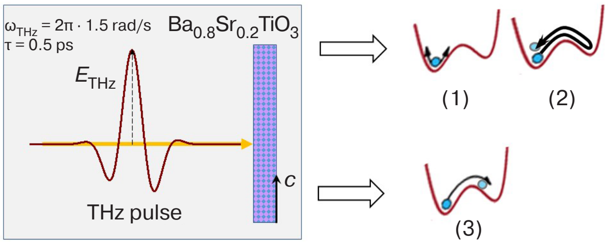


Fig. 1. Schematic diagram of the experiment for which the simulation is performed and a schematic representation of the THz-induced processes of the polarization switching scenarios under consideration

The frequency $\omega_\text{IR}/2\pi$ = 7.55 THz was chosen as the resonant frequency of the IR-active phonon. This corresponds to the frequency of the A_1 (transverse optical) mode [23]. Figure 2 presents the simulation results.

The first calculation was made with varying initial conditions for the IR-active mode (A_IR = 0.1–0.23 Å), while the amplitude of the THz field was insignificant (incapable of causing polarization switching, see the lowermost curve in Fig. 2b). In the calculations, it was assumed that at the initial moment, the total polarization of the cell array under consideration is zero. The other parameters were chosen as follows: γ_IR = 0.5 ps^{-1}, γ_P = 2 ps^{-1}, $C_1 = 5 \cdot 10^{17}$ J/(C·m^2), and $C_2 = 5 \cdot 10^{29}$ J/(C^2·m).

The corresponding time dependencies of the amplitude of the IR-active mode itself and the ferroelectric polarization are shown in Figs. 2a and 2b, respectively. The bottom graph in Fig. 2b presents the time dependence of the THz field (thin maroon line). The insets of Figs. 2b and 2c show the polarization spectra at A_{IR} = 0.23 Å and E_{THz} = 0.8 MV/cm, respectively. The graphs are shifted along the vertical axis for clarity. Dynamic polarization switching occurs already at the initial IR-active mode oscillation amplitude A_{IR} = 0.13 Å. With increasing A_{IR}, dynamic polarization switching is observed in all curves. However, due to the stochastic nature of the solution to the Landau–Khalatnikov equation, given small changes in the variable parameters, permanent switching (i.e., a transition from one of the minima of the potential well to another without a reverse transition at large times) is not possible at all values of the parameters. For example, in Fig. 2, there is no permanent transition at A_{IR} = 0.17 Å.

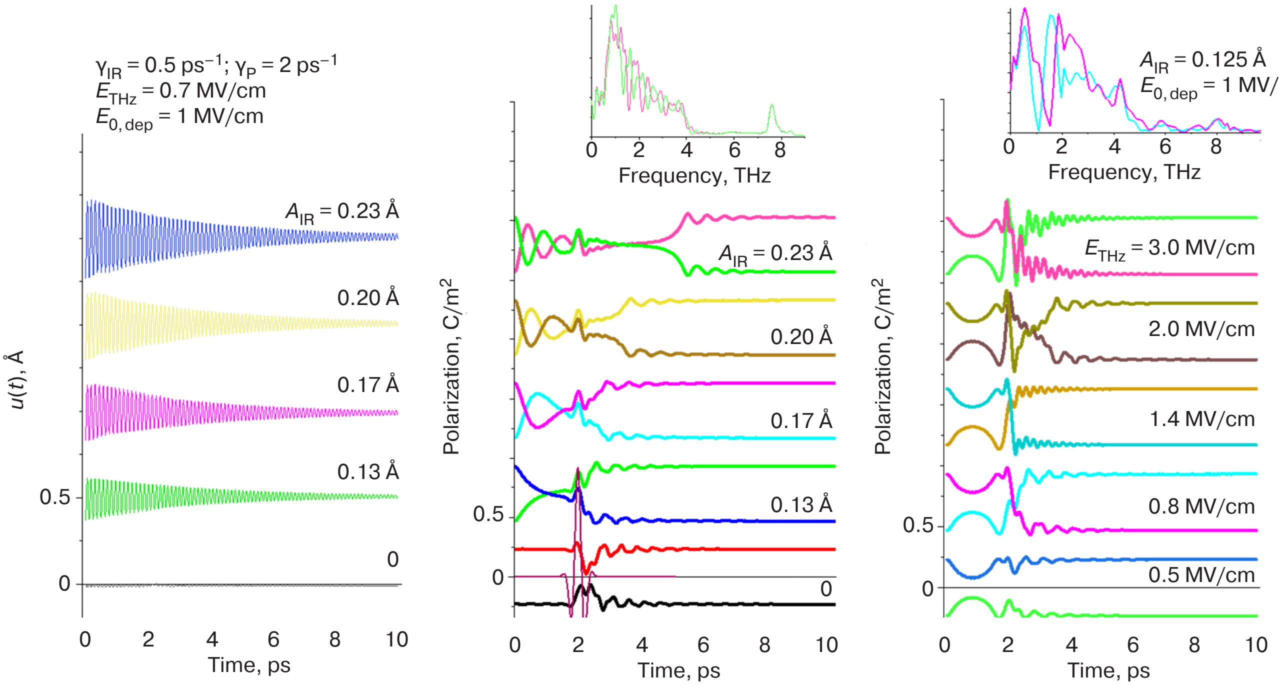
The polarization spectrum (Fig. 2b, inset) exhibits frequencies of both modes: the polar mode in the 1–2 THz range; and the IR-active mode in the 7.55 THz range. Since no features are observed in the time dependencies of polarization at the time delay corresponding to the THz pulse (2 ps), the conclusion can be drawn that the effects observed are associated solely with the action of a coherent optical phonon.

Let us now consider the case of a weak initial IR oscillation (A_{IR} = 0.125 Å), incapable of causing dynamic polarization switching, but with an increase in the THz field. Switching appears at $E_{\mathrm{THz}} \geq 0.8$ MV/cm. The IR-active mode is weakly manifested in the polarization spectra (Fig. 2c, inset). By increasing the THz field amplitude, stable polarization switching is achieved.

Overall, the system of equations considered here does indeed describe polarization switching under the influence of a THz pulse with preliminary excitation of a coherent phonon by a femtosecond optical pulse. The critical effect of the THz pulse, leading to phonon excitation, occurs when the THz pulse has an energy slightly lower (in our calculations, by a maximum of 20%) than the energy required for switching without optical stimulation. Then, the combined effect of these two excitations leads to polarization switching.

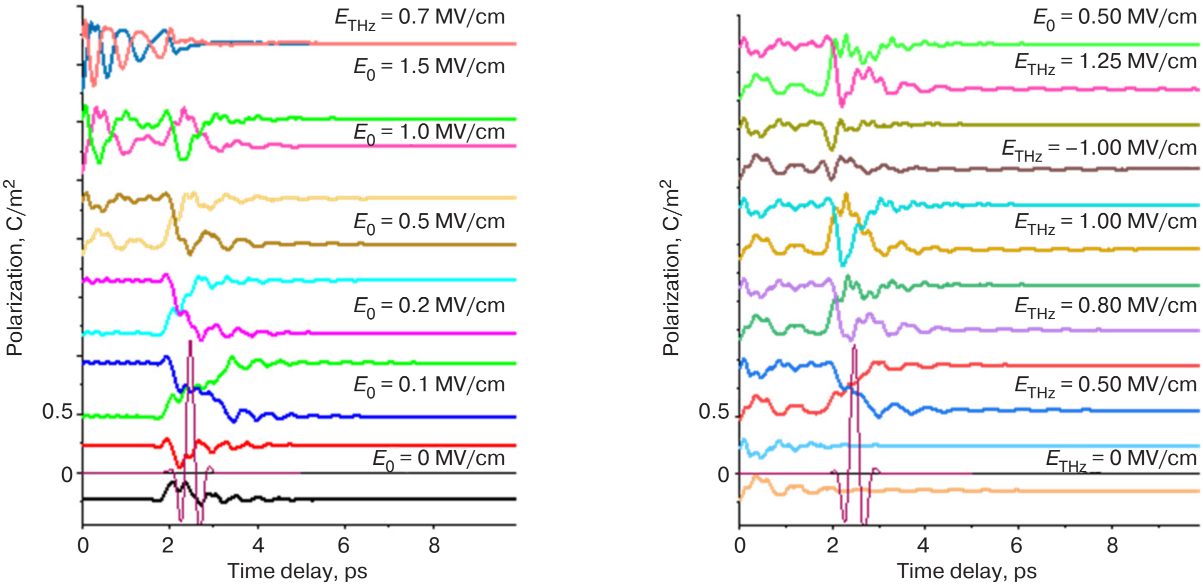
Influence of an additional external electric field

Let us consider the influence of an additional applied electric field E_0 directed parallel to the field E_{THz} of the THz pulse in the plane of the surface of the material. In both cases, the field strengths are chosen in such a way that one field in the absence of the other does not cause polarization switching. In the first case, this condition corresponds to E_{THz} = 0.7 MV/cm (E_0 = 0), while in the second, it corresponds to E_0 = 0.5 MV/cm (E_{THz} = 0). Figure 3 shows the simulation results. The graphs are shifted along the vertical axis for clarity.



(a) (b) (c)

Fig. 2. (a) Time dependencies of the effective coordinate $u(t)$ of the IR-active mode, calculated with varying initial effective amplitude A_{IR} of the IR-active mode at a low field of the THz pulse (the values of the phonon–phonon interaction constants are taken to be equal to $C_1 = 5 \cdot 10^{17}$ J/(C·m^2) and $C_2 = 5 \cdot 10^{29}$ J/(C^2·m)). Time dependencies of the polarization in cells with the opposite initial positions of the effective charge, calculated within the framework of the considered model with varying (b) A_{IR} and (c) the THz pulse field at a small initial amplitude of the IR-active mode



(a)

(b)

Fig. 3. Time dependencies of polarization upon application of an additional constant electric field (a) with increasing strength of the constant field while maintaining the THz pulse amplitude and (b) with increasing THz pulse amplitude while maintaining the strength of the constant field

At the same THz pulse field (Fig. 3a), the constant field with $E_0 \geq 0.1$ MV/cm ($E_0/E_{\mathrm{THz}} = 0.14$) leads to switching of polarization. In this case, also at certain strengths of the constant field ($E_0 = 1$ MV/cm), both ions return to their original state. At fields $E_0 \geq 0.1$ MV/cm, oscillations caused by the constant field alone are observed during the time preceding the THz pulse. The combined action of the constant and THz fields leads to dynamic polarization switching in both cells up to $E_0 = 1.5$ MV/cm ($E_0/E_{\mathrm{THz}} = 2.14$). Further, the THz pulse has practically no effect, and the entire system switches under the influence of a constant field, leading to uniform polarization in the direction of the constant field.

With an increase in the amplitude of the THz field and maintaining the strength of the constant field (Fig. 3b), dynamic switching is observed from $E_{\mathrm{THz}} = 0.5$ mV/cm ($E_0/E_{\mathrm{THz}} = 1$). At some THz field strengths, permanent switching is absent in both branches (0.9 MV/cm < E_{THz} < 1.2 MV/cm), while the behavior of the system at positive values of the constant field (codirectional with the THz pulse field at the point of its maximum amplitude) and its negative values is different. Figure 3b shows the dependencies at $E_0 = -1$ MV/cm, in which even dynamic switching is absent (polarization does not intersect the coordinate axis). A further increase in the THz pulse field leads to the predominance of the response to this pulse, although the constant field has a slight effect on the time dependence of polarization.

All the dependencies presented here are illustrative. The temporal behavior of polarization depends on many parameters (Landau expansion constants and the associated frequency, as well as the soft-mode coefficient).

Nevertheless, an important result of these calculations is the establishment of the fact that even a weak constant field can effectively facilitate polarization switching. This result is extremely important for the experimental observation of polarization switching. Strong THz fields, firstly, are not always achievable and, secondly, can lead to irreversible changes in the physical properties of ferroelectrics [24]. We also noted that the enhancement of the effect of a THz field was previously observed in magnetic materials during studies of magnetization switching in a constant magnetic field.

CONCLUSIONS

This paper proposes a new approach to describing a system with polarization as an order parameter pre-exposed to a femtosecond optical pulse. A phonon mode was excited which subsequently interacted with a high-power THz pulse. The approach is based on a system of the Landau–Khalatnikov equation (where the electric field of the THz pulse and the phonon–phonon interaction act as the driving force) and the phonon mode oscillation equation (where the phonon–phonon interaction acts as the driving force). The novelty of this approach lies in the interaction of the THz pulse with a previously coherently excited phonon mode of significant amplitude.

It was shown that switching occurs at the moment of arrival of a THz pulse, within the time interval of high-amplitude phonon oscillations (determined by the damping constant of this mode). Phonon excitation exerts a critical effect when the THz pulse has an energy slightly lower (in our calculations, by a maximum of 20%) than the energy required for switching without optical stimulation. Then, the combined effect of these two excitations leads to polarization switching. If the field is strong (when polarization can be switched solely by a THz pulse), an increase in the phonon oscillation amplitude does not affect the switching process. Thus, preliminary optical stimulation can induce polarization switching at THz fields lower than the switching threshold. This is crucial for applications, since it allows the use of lower-power THz sources.

The same model considered an additional driving force in the Landau–Khalatnikov equation. It was shown that only low, far-from-saturating, constant fields can effectively promote switching. This result is important for its use in experiments. A negative experimental result had previously been obtained in which the application of a significant field had no effect on polarization switching. The model presented here may explain this result.

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Authors' contribution

All authors contributed equally to the research work.

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