High-frequency polarization switching of a thin ferroelectric film

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We consider both experimentally and analytically the transient oscillatory process that arises when a rapid change in voltage is applied to a $Ba_xSr_{1-x}TiO_3$ ferroelectric thin film deposited on an Mg0 substrate. High frequency ($\approx 10^8$ rad/s) polarization oscillations are observed in the ferroelectric sample. These can be understood using a simple field-polarization model. In particular, we obtain analytic expressions for the oscillation frequency and the decay time of the polarization fluctuation in terms of the material parameters. These estimations agree well with the experimental results.

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I. INTRODUCTION

Unique intrinsic properties make ferroelectric materials attractive both for fundamental research and applications in devices using electro-optical, piezoelectric, and other effects. Bulk ferroelectric materials, particularly those based on barium-strontium titanate $Ba_xSr_{1-x}TiO_3$ (BST) compounds[1,2] are attractive for high-power applications because of their high dielectric permittivity and small losses. Ferroelectric-based devices include ultrafast electrically controlled phase shifters for amplitude and phase control. It has been shown that the dielectric permittivity of a ferroelectric can be altered by applying an electric field. Therefore a fast ferroelectric phase shifter controlled by an electric field bias is being investigated[3] to be used for applications in particle accelerators. Ferroelectric materials provide significant benefits for several applications such as switching and control elements. These are able to handle high peak and average power while maintaining a very short response time of less than just few nanoseconds. According to some estimations[1,3] the response time to an applied external electric field is about 10^{-11} s for crystalline and 10^{-10} s for ceramic compounds. The high permittivity (over 1000) of ferroelectrics makes them potential candidates to replace silicon oxide dielectrics as storage capacitors for memory devices. A solid solution barium-strontium titanate BST has a high permittivity and a composition-dependent Curie temperature T_c which varies in a range of 30–400 K. This strong dependence of the dielectric constant on electric field offers the opportunity to use ferroelectrics as tunable devices. Ferroelectric based phase shifters organized in an array have the advantage of being cheap and consuming a reduced power while continuously tuning the phase of a high-power microwave signal. This rapid electrical steering is realized by adjusting the bias voltages on each element. See, for example Refs. 4 and 5, where the phase shifting elements based on BST thin-film capacitors are discussed. There is a certain advantage in using elements build on thin ferroelectric films because of their compactness and parameter tunability. The presence of internal stresses in the thin film ferroelectrics changes their electromechanical and dielectric properties drastically. Because of this the dielectric permittivity in a thin film is reduced by about an order of magnitude compared to one for the bulk media. However choosing the appropriate substrate allows to adjust the internal stress level and tune the physical properties of the thin ferroelectric films. Thin films of BST deposited by the sputtering technique are discussed in Refs. 6 and 7 in reference to the production of compact tunable capacitors. These elements are attractive for applications in adaptive impedance matching networks and tunable filters. Size effects in thin ferroelectric films are discussed in Ref. 8. It is reported that no critical film thickness is required to obtain polarization switching.

A noteworthy transient effect occurs in ferroelectric material subjected to an alternating electric field which causes polarization switching between two stationary states of the ferroelectric. The switching process is followed by high-frequency polarization oscillations around their stationary states. Basically the polarization dynamics of a ferroelectric near its steady state can be viewed in terms of a damped oscillator with an eigenfrequency determined by the material parameters. An alternating electric field serves as the external force that pushes the oscillator away from its equilibrium. The generation of infrared (IR) radiation by means of polarization switching was proposed for ferroelectrics.[9] There the authors estimate the energy radiated using the dipole approximation.

The study of the transient dynamics of these processes can help understand how the system parameters can be adjusted to provide the required transient behavior. That is important for devices subjected to sharp/shock periodic or aperiodic forces of high frequency. They then spend most of their time in a transient state, even if the relaxation time is smaller than the observation period. Transient processes occurring in ferroelectric might become unwanted effects if the

possible applications require fast switching between polarization states, such as in ferroelectric random access memory. In opposite they might be required if the polarization switching is used to produce oscillations that will form an IR pulse. The polarization relaxation time and oscillation frequency are important characteristics of this transient behavior. The polarization damping constant in ferroelectrics is on the order of 10^{10} s^{-1}, as indicated in Ref. 10. The theoretical description for an isotropic paraelectric in the framework of the dynamical Landau-Khalatnikov model being investigated in Refs. 11 and 12 allows to calculate nonlinear susceptibility coefficients.

Here we discuss an experiment where we observe the polarization dynamics in a thin BST film. A similar experiment studying fast polarization switching was performed earlier by some of the authors. The method of observation of the polarization uses the second harmonic generation in the thin film of the BST solid solution. It is discussed in Ref. 13, where preliminary results were presented. Here we have additional measurements and reinterpret them using a detailed theory. The experiment follows a pump-probe procedure. First we apply a constant field E_s and obtain the steady-state-induced polarization P_s. Then we send in an additional small electric field pulse to probe the film. In principle, this time-dependent solution could be described by the theory set up in Ref. 14 but we chose in this first study to describe the relaxation response of the film. For that we set up a scattering theory formalism. The bound states of the system (E,P) will then give the response of the film, i.e., the typical oscillation frequency and the radiative decay time. We identify two channels of damping, the radiative damping and the inner damping. From the experimental data we estimate the magnitude of both terms and find that here the inner damping dominates. There could be other experiments where the contrary happens, i.e., the radiative damping may dominate.

II. MODEL OF ELECTROMAGNETIC RESPONSE IN FERROELECTRIC FILM

The theory of ferroelectric has started to develop in the 1930s. The phase transition theory proposed by Landau[15,16] was applied to describe the behavior of the ferroelectric near a critical point of the phase transition. Following the Landau-Ginzburg-Devonshire theory the thermodynamic Gibbs potential Φ in the neighborhood of the critical point can be represented as a power series of the order parameter, namely, the polarization P,[17–20]

$$\Phi = \Phi_0 + \frac{\alpha}{2}P^2 + \frac{\beta}{4}P^4 - EP.$$

We have considered that the film is made of an isotropic material for simplicity. Anisotropic effects can be principal for bulk materials because they could accumulate. This is not the case for a thin film. The expansion coefficients are $\alpha = a_0(T - T_c)$ and β. T_c is the Curie temperature. In a solid solution $Ba_xSr_{1-x}TiO_3$ the Curie temperature depends on the relative concentration x of barium. The approximate formula to calculate T_c in BST was proposed by Ref. 21. It reads $T_c = 360x + 40$. Particularly $T_c = 292$ K for $x = 0.7$. This is the concentration of barium in the thin film investigated in an experiment on ferroelectric switching.[13]

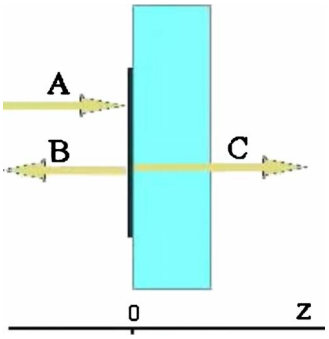

FIG. 1. (Color online) Schematics of the experimental setup where an electric field pulse polarizes a ferroelectric thin film deposited on a substrate.

We describe an experiment where a thin film of thickness l is deposited on a dispersionless substrate and is submitted to an incident electric field as shown in Fig. 1. The optical properties of the surrounding media can be characterized by the refractive index $n(z)$, such that $n(z) = 1$ for $z < 0$ and $n(z) = n > 1$ for $z > 0$, i.e., in the substrate. The electric field $E(z,t)$ is governed by the Maxwell equation and the polarization obeys the equation of a damped oscillator driven by the field (compare to Ref. 14)

$$\frac{\partial^2 E}{\partial z^2} - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{l}{\epsilon_0 c^2}\frac{\partial^2 P}{\partial t^2}\delta(z),$$

$$\frac{\tau^2}{\epsilon_0}\frac{\partial^2 P}{\partial t^2} + \frac{\gamma\tau^2}{\epsilon_0}\frac{\partial P}{\partial t} + \alpha P + \beta P^3 = E_f(t), \tag{1}$$

where $E_f(t)$ is the electric field inside the thin film. The constant τ is proportional to the inverse of the Born frequency, this value will be estimated for BST later. Typically the decay constant $1/(\gamma\tau^2)$ is small.[19] However we take it into account for generality.

The electromagnetic wave is incident from the left of a film located at $z = 0$. Thus the electric fields outside the thin film $E^-(z,t)$ for $z < 0$ and $E^+(z,t)$ for $z > 0$ are defined by free wave equations. The time Fourier images of these fields can be written as

$$\tilde{E}^-(z,\omega) = Ae^{ik_1 z} + Be^{-ik_1 z},$$

$$\tilde{E}^+(z,\omega) = Ce^{ik_2 z}.$$

Here $k_1 = \omega/c$ and $k_2 = \omega n/c$ are the wave numbers on the left and on the right of the thin film, A is the Fourier amplitude of the incident wave, B is the Fourier amplitude of the reflected wave, and C is the Fourier amplitude of the transmitted wave.

According to the boundary conditions at $z = 0$ (Refs. 14 and 22) the electric field $\tilde{E}$ and its spatial derivative $\tilde{E}_{,z}$ are connected by the relations

$$\tilde{E}^{(-)}(z = 0,\omega) = \tilde{E}^{(+)}(z = 0,\omega),$$

$$\tilde{E}_{,z}^{(-)}(z=0,\omega)-\tilde{E}_{,z}^{(+)}(z=0,\omega)=\frac{l\omega^2}{c^2\epsilon_0}P(\omega),$$

where $P(\omega)$ is the Fourier image of the thin-film polarization. This leads to the following equations for the Fourier amplitudes of the left and right electric fields

$$A+B=C,\quad A-B=nC-\frac{il\omega}{c\epsilon_0}\tilde{P}(\omega).$$

Hence the amplitude of the transmitted wave C and the amplitude of the reflected wave B can be expressed through the amplitude A of the incident wave and the thin-film polarization $P(\omega)$

$$C=\frac{2}{n+1}A+\frac{il\omega}{c(n+1)\epsilon_0}\tilde{P}(\omega), \tag{2}$$

$$B=\frac{1-n}{1+n}A+\frac{il\omega}{c(n+1)\epsilon_0}\tilde{P}(\omega). \tag{3}$$

Using the inverse Fourier transform we obtain the amplitude of the electric field inside the thin film E_f as $E^+(z,t)$ at $z=0$,[22] i.e.,

$$E_f(t)=\frac{2}{1+n}E_{in}(t)-\frac{l}{c(n+1)\epsilon_0}\frac{\partial P}{\partial t},$$

where $E_{in}(t)$ is the electric field of the incident wave. Thus one can find from the second equation of the system in Eq. (1) that the polarization of the ferroelectric thin film is governed by the following equation:

$$\frac{\tau^2}{\epsilon_0}\frac{\partial^2 P}{\partial^2 t}+\left[\frac{\gamma\tau^2}{\epsilon_0}+\frac{l}{c(n+1)\epsilon_0}\right]\frac{\partial P}{\partial t}+\alpha P+\beta P^3=\frac{2}{1+n}E_{in}(t). \tag{4}$$

This expression shows that two relaxation channels exist. One of them is the ordinary one related with inner friction, i.e., the γ term. The second relaxation channel is due to the radiation process. The variation in the polarization generates the electromagnetic field outside the film.

The polarization of the thin film evolves according to Eq. (4) as an oscillator with damping and a forcing that is equal to $E_s=2E_{in}(t)/(1+n)$. Furthermore, this equation allows to consider E_{in} as a constant electric field, i.e., a constant voltage. In this case $E_s=V_s/l$, where V_s is the applied voltage.

A. Relaxation to a steady-state polarization

In the absence of an external electric field the ferroelectric possesses two equilibrium states, each corresponding to different polarities. In the paraelectric phase there is only one equilibrium corresponding to an unpolarized state. The experiment under consideration was performed at room temperature which is above the Curie temperature for BST so that the sample is in the paraelectric state. However there is another way to change the polarization state. This can be induced by applying an external field or a stress. In this experiment we chose to do the former. The induced steady-state polarization P_s is defined as the fixed point of the Eq. (1), where we assume that the electric field $E_f(t)=E_s$ is constant and the polarization $P=P_s$ are time independent. We get

$$\alpha P_s+\beta P_s^3=E_s. \tag{5}$$

We assume the solution of Eq. (1) to be of the form

$$E=E_s+\delta E,\quad P=P_s+\delta P, \tag{6}$$

where δE and δP are small compared, respectively, to E_s P_s. Plugging this into the Eq. (1) we get the linearized equations around the stationary solution (E_s,P_s)

$$\frac{n^2}{c^2}\frac{\partial^2\delta E}{\partial t^2}-\frac{\partial^2\delta E}{\partial z^2}=-\frac{l}{\epsilon_0 c^2}\frac{\partial^2\delta P}{\partial t^2}\delta(z),$$

$$\frac{\tau^2}{\epsilon_0}\frac{\partial^2\delta P}{\partial t^2}+\frac{\gamma\tau^2}{\epsilon_0}\frac{\partial\delta P}{\partial t}+(\alpha+3\beta P_s^2)\delta P=\delta E(t). \tag{7}$$

B. Scattering of linear waves by the thin film

We now proceed to solve the linearized Eq. (7) by using a scattering theory formalism. We separate time and space by assuming a periodic solution

$$\delta E=e(z)e^{-i\omega t},\quad \delta P=pe^{-i\omega t}. \tag{8}$$

We get

$$n^2k^2e+\frac{\partial^2 e}{\partial z^2}=-\frac{l}{\epsilon_0}k^2\delta(z)p,$$

$$(\Omega^2-i\gamma\tau^2\omega-\tau^2\omega^2)p=\epsilon_0 e(0), \tag{9}$$

where the wave number $k=\omega/c$ and where we introduced

$$\Omega^2=\epsilon_0(\alpha+3\beta P_s^2). \tag{10}$$

In the scattering we assume the electromagnetic wave to be incident from the left of the film located at $z=0$. We then have

$$e=e^{ikz}+Re^{-ikz},\quad z<0;\quad e=Te^{iknz},\quad z>0, \tag{11}$$

where R is the amplitude of the reflected wave and T the amplitude of the transmitted wave. These expressions are related to the A, B, C parameters of the previous section through the relations $R=B/A$, $T=C/A$. We have the following interface conditions at $z=0$:[14,22]

$$e(0^-)=e(0^+),\quad -[e_z]_{0^-}^{0^+}=k^2\frac{l}{\epsilon_0}p. \tag{12}$$

They imply

$$1+R=T,\quad nT-(1-R)=-ik\frac{l}{\epsilon_0}p.$$

Using the second relation of Eq. (9) to obtain p we get the transmission coefficient T

$$T=\frac{2c(\Omega^2-i\gamma\tau^2\omega-\tau^2\omega^2)}{c(n+1)(\Omega^2-i\gamma\tau^2\omega-\tau^2\omega^2)+il\omega}. \tag{13}$$

The refraction coefficient is

$$R = \frac{c(1-n)(\Omega^2 - i\gamma\tau^2\omega - \tau^2\omega^2) - il\omega}{c(1+n)(\Omega^2 - i\gamma\tau^2\omega - \tau^2\omega^2) + il\omega}. \tag{14}$$

The bound states are the poles of the reflexion and transmission coefficients. Their existence indicates that the system has resonant modes that can be excited by an incoming wave. The real part of the bound states is the oscillation frequency and the imaginary part is the inverse of the decay time of the mode.

The poles of R, T are given by

$$c(n+1)(\Omega^2 - i\gamma\tau^2\omega - \tau^2\omega^2) + il\omega = 0,$$

which is the second degree equation

$$\omega^2\tau^2 - i\omega\left[\frac{l}{(n+1)c} - \gamma\tau^2\right] - \Omega^2 = 0, \tag{15}$$

whose roots are

$$\omega = \frac{i}{2}\left[\frac{l}{(n+1)c} - \gamma\right] \mp \sqrt{\frac{\Omega^2}{\tau^2} - \frac{1}{4}\left[\frac{l}{(n+1)c} - \gamma\right]^2}. \tag{16}$$

The imaginary part and real part of $\omega = \omega_r + i\omega_i$ give, respectively, the decay time T_{dec} and the oscillation period T_{osc}

$$T_{dec} = \frac{1}{\omega_i}, \quad T_{osc} = \frac{2\pi}{\omega_r}. \tag{17}$$

We will estimate these parameters for the experiment in the next section.

C. Green's function solution of the linearized equation

The linearized equation

$$\frac{\tau^2}{\epsilon_0}\frac{\partial^2\delta P}{\partial t^2} + \frac{\tau^2\gamma}{\epsilon_0}\frac{\partial\delta P}{\partial t} + (\alpha + 3\beta P_s^2)\delta P = \delta E(t)$$

can be solved to obtain the polarization response $\delta P(t)$ to a give incoming electric field $\delta E(t)$. For that we introduce the Green's function $G(t-t_0)$ which satisfies

$$\tau^2\frac{\partial^2 G}{\partial t^2} + \tau^2\gamma\frac{\partial G}{\partial t} + \Omega^2 G = \epsilon_0\delta(t-t_0). \tag{18}$$

Using the Laplace transform

$$\hat{G}(s) \equiv \int_0^\infty e^{-st}G(t)dt$$

and assuming that $G(0)=0$ and $\partial G/\partial t(0)=0$ we obtain

$$\hat{G}(s) = \frac{\epsilon_0 e^{-st_0}}{\tau^2 s^2 - \tau^2\gamma s + \Omega^2}. \tag{19}$$

To get the inverse Laplace transform one expands this rational function as

$$\hat{G}(s) = \frac{\epsilon_0}{2(s_1 - s_2)}\left(\frac{e^{-st_0}}{s - s_1} - \frac{e^{-st_0}}{s - s_2}\right),$$

where s_1, s_2 are the roots of the denominator of Eq. (19). This yields the Green's function

$$G(t-t_0) = \frac{\epsilon_0}{2(s_1 - s_2)}[e^{s_1(t-t_0)} - e^{s_1(t-t_0)}], \quad t > t_0,$$

$$G(t-t_0) = 0, \quad t < t_0.$$

The roots are complex conjugate $s_1 = \omega_i + i\omega_r$, $s_2 = \omega_i - i\omega_r$ so we obtain the final result

$$G(t-t_0) = e^{\omega_i t}\frac{\sin[\omega_r(t-t_0)]}{2\omega_r}, \quad t > t_0,$$

$$G(t-t_0) = 0 \quad t < t_0. \tag{20}$$

The polarization response $\delta P(t)$ to a given perturbation of the electric field $\delta E(t)$ is the convolution integral

$$\delta P(t) = \int_{-\infty}^{t} G(t-t_0)\delta E(t_0)dt_0. \tag{21}$$

D. Induced polarization caused by a short electric pulse

Now let us consider an alternative method to investigate the polarization response. Suppose that a ferroelectric film is in a polarized state P_s caused by a constant electric field E_s. At certain moment we send in a short electric pulse so the polarization of the film changes during a short time period. After that the polarization relaxes to the steady-state position P_s. If the electric pulse is sufficiently short, i.e., close to a "δ function" then the polarization response will follow the Green's function in Eq. (20). We term this action "δ-functionlike pushing" the nonequilibrium polarization.

The linearization of the equation for the polarization in Eq. (4) near the steady state P_s assuming $P = P_s + p$ with the condition $p \ll P_s$ results in the equation for the Green's function in Eq. (18). Let us suppose that the extremely short electric pulse acts at $t=0$. Then we can conclude that the evolution of $p(t)$ after $t=0$ is described by

$$p(t) \sim e^{\omega_i t}\frac{\sin(\omega_r t)}{2\omega_r}.$$

Hence the polarization decay rate is defined by the imaginary part of the complex frequency in Eq. (16), ω_i and the corresponding time for the polarization to attain the equilibrium is

$$T_{dec} = 1/\omega_i.$$

The real part of the frequency in Eq. (16) corresponds to oscillations of the polarization as it is approaches the steady-state value. Here we recover the results obtained using the scattering formalism.

III. EXPERIMENTAL RESULTS

The experiment was performed using the nonlinear optical stroboscopic technique as in Ref. 13. For the second harmonic (SH) generation, the radiation of a titanium-sapphire laser (MaiTai, New-Port-SpectraPhysics) was used with a pulse duration of 100 fs, a wavelength of 780 nm, a repetition rate of 100 MHz, and an average power of 100 W. The

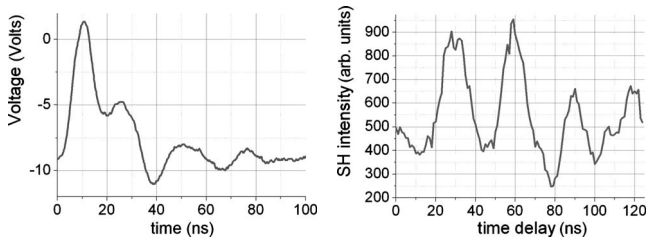

FIG. 2. Plots of a positive "zero" electric pulse voltage as a function of time (left panel) and the subsequent polarization oscillations of the film as a function of time (right panel). The constant field E_s is negative.

experiment was performed at room temperature. The 70-nm-thick $Ba_{0.7}Sr_{0.3}TiO_3$ films were deposited onto a MgO substrate by rf sputtering. For such a composition, the Curie temperature equals $T_c=20$ C. However for thin films the phase transition is blurred around this value and this is confirmed by the presence of a narrow hysteresis above T_c.[13] A voltage pulse of duration about 25 ns, produced by an Avtech pulse generator, was applied to the copper contacts on the BST film. The polarization response of the ferroelectric film was measured as the coherent SH intensity in the experiment.[23]

Figures 2 and 3 show the polarization response as a function of time of the BST thin film to electric pulses having the same amplitudes and opposite polarities. The left panels show the incoming electric field pulses as a function of time and the right panels show the SH intensity (proportional to the square of the polarization) as a function of time. In Fig. 2 the electric field pulse (left panel in the figure) is realized by a rapid spiking to a zero value from a constant negative voltage background with return to the same constant negative value. For Fig. 3 the electric pulse (left panel at the figure) drops from its constant positive value to zero and returns to the constant. In both measurements the spiking pulse will be called the 'zero' pulse. The pulse profile is intended to have a narrow bell shape, however because of certain setup drawbacks a low-amplitude tail appears. In the right panels of Figs. 2 and 3 it can be seen that the polarization oscillates long after the "zero" electric pulse has passed through the thin film. Then the eigenfrequency of the polarized thin film can be determined.

In both pictures the polarization oscillates around its stationary values (defined by the constant background electric field) with very close oscillations periods, about 60 ns. This agrees with the relation in Eq. (22) in the limit when the

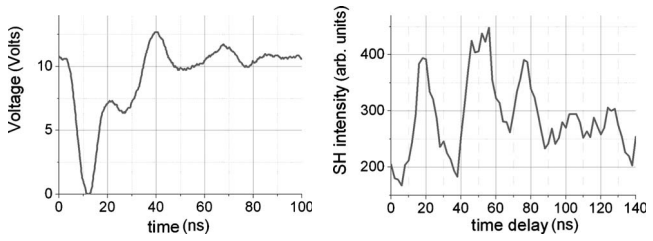

FIG. 3. Plots of a negative "zero" electric pulse voltage and the subsequent polarization oscillations of the film as in Fig. 2 except that the constant field E_s is positive.

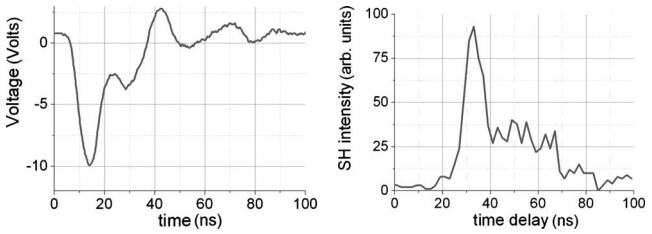

FIG. 4. Plots of a "normal" electric pulse voltage and the subsequent polarization oscillations.

second term can be neglected in the square root because Ω only depends on the amplitude $|E_s|$.

The next Fig. 4 shows the polarization dynamics when there is no constant electric field E_s after a delta function such as electric pulse of negative polarity (shown in the left panel) passes through the film. This type of electric pulse is the analog of a "normal pulse" studied experimentally in Ref. 13. When the film is not polarized the SH cannot be generated so that its intensity is about zero. The SH signal from the perturbed unpolarized state is a few times smaller than the one for the previous experiments. There the steady polarized state of the thin film was studied by a zero pulse. There are almost no polarization oscillations when perturbing a nonpolarized thin film. Again this agrees with the estimate in Eq. (22).

Discussion of the experimental results

There is a relatively broad range of experimentally obtained values of the Landau coefficients in BST solid solutions. The numbers vary with the fabrication method. The Landau coefficients found by[24] for a BST solid solution $Ba_{0.7}Sr_{0.3}TiO_3$ in SI units are presented in Table I. In the experiment performed the film thickness is $l=70$ nm (as in Ref. 13). We assume that the substrate refraction index is $n=1.5$. These parameters enable to calculate the frequency Ω. For a normal pulse the applied voltage $V_s=0$ so $E_s=0$ and $P_s=0$. This gives $\Omega^2=\epsilon_0\alpha\approx 2\times 10^{-5}$. In the presence of an electric field $E_s=V_s/l$ where $V_s=10$ V we have $E_s\approx 10^8$ V/m so that from Eq. (5) we get $P_s\approx 0.79$ C/m^2. This gives $\Omega^2=\epsilon_0\alpha\approx 1.8\times 10^{-3}$.

First let us estimate the term $l[(n+1)c]^{-1}$ in the formula (16). We have $l[(n+1)c]^{-1}\approx 10^{-16}$ which is very small so that this radiative damping can be completely neglected for this particular experimental situation. From the experimental data it can be seen that the polarization response for a voltage $V_s=0$ is qualitatively different from the one for $V_s=10$ volts. In particular, it has a smaller decay time and practically no oscillations. Let us estimate the decay time T_{dec} from Eq. (16). We have

TABLE I. Material parameters for $Ba_{0.7}Sr_{0.3}TiO_3$.

T_c (C)	α (m/F)	β (m^5/C^{2}F)
34	2.2×10^6	2.52×10^8

TABLE II. The decay time T_{dec} as a function of the parameters (τ, γ) for the zero pulse $V_s=0$ V.

	$\gamma=10^6$ (s^{-1})	10^7	10^8	10^9
$\tau=10^{-6}$ [s]	10^{-6}	10^{-7}	10^{-8}	10^{-9}
10^{-7}	10^{-6}	10^{-7}	10^{-8}	10^{-9}
10^{-8}	1.4×10^{-6}	10^{-7}	10^{-8}	10^{-9}
10^{-9}	2×10^{-6}	1.4×10^{-7}	10^{-8}	10^{-9}
10^{-10}	2×10^{-6}	2×10^{-7}	1.4×10^{-8}	10^{-9}
10^{-11}	2×10^{-6}	2×10^{-7}	2×10^{-8}	1.4×10^{-9}
10^{-12}	2×10^{-6}	2×10^{-7}	2×10^{-7}	2×10^{-9}

$$T_{dec} = 2/\gamma$$

if $(\Omega^2/\tau^2 - \gamma^2/4) > 0$, otherwise

$$T_{dec} = \left(\frac{\gamma}{2} + \sqrt{\frac{\gamma^2}{4} - \frac{\Omega^2}{\tau^2}}\right)^{-1}.$$

This decay time is shown in Table II for different values of the free parameters (τ, γ). Clearly if $(\Omega^2/\tau^2 - \gamma^2/4) > 0$ there will be oscillations in the polarization. Comparing the estimates of the table with the result shown in Fig. 4 indicates that $\gamma \approx 10^7$ s^{-1}. This agrees with the value obtained in a previous study by the authors.[23]

Let us now compare the oscillation period obtained for the nonzero pulse $V_s=10$ volts with the estimate in Eq. (16). We have

$$T_{osc} = 2\pi\left(\sqrt{\frac{\Omega^2}{\tau^2} - \frac{\gamma^2}{4}}\right)^{-1}. \tag{22}$$

This value is computed for different values of γ and τ and reported in Table III. The zero entries correspond to the situation where $\Omega^2/\tau^2 - \gamma^2/4 < 0$. As can be seen the values closest to what is observed in the experimental plots Figs. 2 and 3 are $\tau=10^{-10}$ s and $\gamma=10^7$. This value of the oscillation time τ agrees with the one that can be computed using the speed of sound $c_s=510^3$ m/s in the material and the film thickness l. We get $\tau=l/c_s \approx 10^{-11}$ s. The estimate for the bulk material using the Newton equation describing the lattice oscillations $\tau_{hf}^2 \approx M\epsilon_0/(Ne^2) \approx 10^{-26}$ s^{-2}, where M is the atomic mass of BST gives $\tau \approx 10^{-13}$ s which is much too small.

TABLE III. The oscillation period T_{osc} as a function of the parameters (τ, γ) for the nonzero pulse $V_s=10$ V.

	$\gamma=10^6$ (s^{-1})	10^7	10^8	10^9
$\tau=10^{-6}$ [s]	0	0	0	0
10^{-7}	1.5×10^{-5}	0	0	0
10^{-8}	9.6×10^{-7}	1.5×10^{-6}	0	0
10^{-9}	9.6×10^{-8}	9.6×10^{-8}	1.5×10^{-7}	0
10^{-10}	9.6×10^{-9}	9.6×10^{-9}	9.6×10^{-9}	1.5×10^{-8}
10^{-11}	9.6×10^{-10}	9.6×10^{-10}	9.6×10^{-10}	9.6×10^{-10}
10^{-12}	9.6×10^{-11}	9.6×10^{-11}	9.6×10^{-11}	9.6×10^{-11}

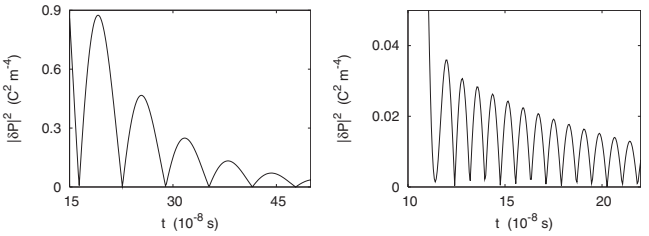

FIG. 5. The polarization response of the film for a normal pulse $(E_s=0)$ (left panel) and for a zero pulse (right panel).

We have used the expression (21) to compute the polarization response of the system to a given perturbation δE of the electric field. We assume a Gaussian δE

$$\delta E(t) = \exp\left(-\frac{t^2}{2w_e}\right) \tag{23}$$

and normalize times by $T=10$ ns. We chose $\gamma=10^7$ s^{-1}, $\tau=10^{-10}$ s. In the normalized units we have $\gamma=0.1$, $w_e=1$. The normalized frequency $\omega=0.45$ for the zero pulse and $\omega=4.5$ for the normal pulse. We calculated the polarization response by numerical integration using the trapeze method. To compare with the experimental data we computed δP^2. The results are presented in Fig. 5 for a normal pulse $(E_s=0)$ in the left panel and for a zero pulse in the right panel. One can see on the left side of the plots the first response of the polarization. We have omitted it because it is the forced response due to the probing pulse δE. We only present the subsequent free evolution of δP. For $V_s=0$ the polarization decays with few oscillations. For $V_s=10$ V the polarization oscillates much longer. This is in quantitative agreement with the experimental plots presented above.

Finally let us consider the important issue of how much heat the sample receives during its irradiation by the laser. We assume that the film is a dielectric, thus the Joule heat is negligible. Our model allows to predict how much heat is absorbed by the dielectric material. The permittivity $\varepsilon(\omega)$ has an imaginary part $\varepsilon''(\omega)$ which depends on the frequency ω. This is due to the Causality principle. The heat absorbed per unit volume is defined by the following formula:

$$q(\omega) = \frac{\omega}{8\pi}\varepsilon''(\omega)|e|^2,$$

where ω is the carrier frequency of the electromagnetic wave and e is the amplitude of this wave. We consider a harmonic wave. We can find $\varepsilon''(\omega)$ in the framework of our model. From Eq. (9) we can obtain the polarization of unit volume p as

$$p = \frac{\epsilon_0 e(0)}{\Omega^2 - \tau^2\omega^2 - i\tau^2\gamma\omega}.$$

The polarization of matter in the volume lS (S being the square section of the beam) is plS. Using the expression $D=\epsilon_0 e(0) + plS = \epsilon_0 \varepsilon e(0)$, one can find the *permittivity* $\varepsilon(\omega)$ as

$$\varepsilon(\omega) = 1 + \frac{lS}{\Omega^2 - \tau^2\omega^2 - i\tau^2\gamma\omega}. \quad (24)$$

This formula gives the imaginary part of $\varepsilon(\omega)$

$$\varepsilon''(\omega) = \frac{lS(\tau^2\gamma\omega)}{(\Omega^2 - \tau^2\omega^2)^2 + (\tau^2\gamma\omega)^2}.$$

Assuming the following values of the parameters:

$$\Omega^2 = 1.8 \times 10^{-3}, \quad \gamma = 10^7 \text{ s}^{-1}, \quad \tau = 10^{-10} \text{ s}, \quad l = 7 \times 10^{-8} \text{ m},$$

we have

$$\varepsilon''(\omega) \approx \frac{7 \times 10^{-21} S\omega}{(1.8 \times 10^{-3} - 10^{-20}\omega^2)^2 + 10^{-26}\omega^2}.$$

Let the frequency ω correspond to the light diapason, i.e., $\omega = 10^{15}$ s^{-1}. In this case $\varepsilon''(\omega) \approx 7 \times 10^{-26} S$. The corresponding heat is equal to

$$q \approx 0.3 \times 10^{-11} S|e_0|^2.$$

We should take a very small intensity of probe wave in the case of the spectroscopic (scattering) technique. The transverse section S is about 10^{-5} m^2. Thus in this case the heating effect is extremely small. In the second-harmonic generation (SHG) experiment $|e_0|^2 \sim 10-10^2$ so $q \ll 1$ in this case too. This confirms the noninvasive nature of the the SHG technique because the resonant frequency of the polarization response is much smaller than the laser frequency. If we choose a laser frequency that is close to the resonant frequency $\omega = \Omega/\tau$ then $\varepsilon''(\omega)$ is given by

$$\varepsilon''(\omega) \approx \frac{lS}{\gamma\tau\Omega}.$$

Using the same parameters as above we can estimate this expression as $\varepsilon''(\omega) \approx 4 \times 10^{-2} S$ The heat $q = 10^6 S|e_0|^2$ which is significant even for fairly small fields e_0. The SHG technique now strongly perturbs the sample.

IV. CONCLUSION

We analyzed the polarization oscillations occurring in a thin ferroelectric film as a short electric pulse crosses it. We consider that the ferroelectric is in the paraelectric phase (high temperature). As in a pump probe experiment the film is initially in a static polarization state P_s induced by a constant voltage V_s. Using the Landau-Ginzburg-Devonshire theory we computed this static polarization and the subsequent oscillations of the polarization induced by a short voltage pulse. These were analyzed using a scattering theory formalism and a Green's function approach. Two channels of dissipation were identified, a radiative damping and an intrinsic damping.

This theory was applied to explain the experimental time evolution of the polarization for a thin ferroelectric film of BST. The polarization is estimated indirectly through second harmonic generation. For this experimental situation we show that the radiative damping can be neglected and only the intrinsic damping should be considered. From a comparison of our model to the experimental plots we estimated the important parameters τ the response time and γ the relaxation coefficient. Using these values our theoretical estimates of the decay time and of the oscillation period agree well with the observations. In particular, for a normal pulse for which $V_s = 0$ the damping dominates and we only see a few oscillations of the polarization. On the contrary for a zero pulse for which $V_s = 10$ V the damping time is longer compared with the oscillation period. Here we obtain many oscillations of the polarization.

Although the radiative damping appears as artificial in this particular high-temperature situation, at low-temperature phonons become frozen and the radiative damping may become predominant. The model can be further elaborated by including the spatial inhomogeneity of the thin film, the depolarization effects of the boundaries, and taking into account the internal stresses of the film. Also the dynamics of the polarization of a thin-film multilayered structure can be investigated by generalizing the model.

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