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# Dynamical magnetoelectric effects associated with ferroelectric domain walls

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## Abstract

Molecular dynamics simulations using a first-principles-derived effective Hamiltonian are conducted on lead zirconium titanate ultrathin films possessing nanoscale ferroelectric domains and being under GHz electric field. Pulses of magnetization are predicted to occur in this system, when sudden changes of morphology of these nano domains occur. A simple equation relating the magnetization and product between the electrical polarization and its time derivative is found to reproduce and explain these magnetization pulses, as well as previously observed magnetoelectric effects in *moving* ferroelectric domain walls/phase boundaries in ferroelectrics and magnetoelectrics.

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The effect of a magnetic field on phase boundary of ferroelectrics has been known for around 30 years, since Flerova and Bochkov succeeded in orienting the phase boundary separating the ferroelectric and paraelectric phases of a $BaTiO_3$ lamella by cooling under a magnetic field [1, 2]. Once equilibrium is reached under such cooling, the electrical polarization of the ferroelectric phase was always found to lie along a direction (say, the $z$ axis) that is perpendicular to the applied magnetic field (that is oriented, say, along the $y$ axis) with these two orthogonal vectors defining the phase boundary plane. Interestingly, this control of the phase boundary by a magnetic field was only found, when this boundary was moving along the third, perpendicular axis (the $x$ axis). In particular, the phase boundary was unresponsive to the application of the magnetic field in the stationary state. Similarly, Popov, Tikhomirova, and Flerova [3] found that the magnetic field can also influence the orientation of the ferroelectric domain walls (FDW) in nonmagnetic gadolinium molybdate $Gd_2(MoO_4)_3$. Once again, the control of this orientation was only observed, when the FDW were moving (via the application of rectangular pulses of the electric field [3]), and the geometry of the set-up is well defined: the plane of the FDW is spanned by the direction of the electric dipoles and the applied magnetic field (that are perpendicular to each other) and the FDW is moving along its normal. Interestingly, other *dynamical* magnetoelectric (ME) effects have also been reported. This includes the measurement of a magnetization in gadolinium molybdate [4], as a result of the motion of 180-degree FDW that was initiated by the application of pulses of electric field.

Different explanations of the aforementioned phenomena have been advocated [4, 5] about 20 or 30 years ago. For instance, Orlov *et al* [4] modeled the FDW as a finite slab of width $\lambda$, height $d$ and depth $l$, and assumed that the motion of FDW along the axis defining the $\lambda$ width results in a *local* closed loop of the electric current flowing in this slab. The current density $j = \partial \mathcal{P}/\partial t$ was further assumed to be of constant magnitude within the slab (note that $\mathcal{P}$ is the polarization field, which spatial average is the macroscopic polarization, and which has different sign in the nearest domains separated by the FDW). Such assumptions resulted in the prediction of a finite magnetic moment given by $\mu = P_0 v d l \lambda$, where $P_0$ is the magnitude of the polarization in the domains and $v$ is the velocity at which the domains move, since electrodynamics indicates that a loop of electric current $I$ with cross-section $A$ produces a magnetic moment of magnitude $\mu = I \cdot A$. On the other hand, Flerova and Chupis considered another model, based on the translational movement of a topological soliton (in the form of FDW) [5]. They proposed that the polarization profile of the FDW in gadolinium molibdate is not symmetric with respect to the center of FDW, and, as a result, the FDW motion is accompanied by the displacement of a polarized part of the sample, having the size of the FDW. Such latter displacement creates, according to Lorentz transformation [6], a magnetization $\mathbf{M} = \left[\bar{\mathbf{P}} \times \mathbf{v}\right]$, where $\mathbf{v}$ is the velocity of the FDW and $\bar{\mathbf{P}}$ is the polarization of this polarized part.

Based on these interesting results, it is legitimate to determine if the FDW, separating *nanoscale* domains recently observed in utrathin films [7], can also exhibit such ME effects, when moving, and what is the effect on the (hypothetical) magnetization of the ability of FDW to dramatically change their morphology under applied electric field [8, 9]. Such determination would be timely and important, once realizing that ME effects, dynamics, nano science, and FDW are all topics of current interest [10–13]. It is also worthwhile to know if the assumptions of the models of Refs. [4, 5] are valid in these ultrathin films or, rather, if other models are to be developed.

The goal of this Letter is to resolve the aforementioned issues. For that, we perform first-principles-based molecular dynamics simulations on Pb(Zr,Ti)$O_3$ (PZT) ultra thin films being under an *ac* electric field and possessing up and down nanoscale domains. Based on the recently discovered relationship between the time derivative of the electric toroidal moment and magnetization [14], it is indeed predicted that a magnetization should occur in these systems. This magnetization has two different features and microscopic origins: it can be due to the change of magnitude of electric dipoles within non-moving "up" and "down" domains, and can also exhibit pulses when domains move via sudden change of their morphology. A simple Equation, that directly relates the electric toroidal moment and the product between the polarization and its time derivative, nicely explains the existence of these magnetic pulses in our ultra thin films, as well as the previous observation of Refs. [1–4] on thicker systems.

Here, we consider (001) PZT epitaxial thin films with a Ti composition of 60%, which leads to a ferroelectric tetragonal ground-state in the bulk, and select a $30 \times 4 \times 20$ supercell that is periodic along the $x$ and $y$ directions but finite along the $z$ axis. We use the Molecular Dynamics (MD) approach implemented in previous studies (see, e.g., Ref. [15]) with the Newtonian equations of motion solved for the degrees of freedom of the effective Hamiltonian described in the Supplemental Material [16–23] (see [24]). It is important to realize that such effective Hamiltonian does not incorporate any magnetic degrees of freedom, as consistent with the fact that none of the ions in PZT possesses local magnetic dipoles. An Evans-Hoover thermostat [25–27] is employed in these simulations, in order to equilibrate the system at a fixed temperature of 10 K. At first, we equilibrate the given PZT film under no electric or/and magnetic field by using 200,000 MD time steps, each being 0.5 fs long. Then, we apply, along the $z$ axis, the combination of a bias and an ac electric fields $E(t) = 2.5[1 - sin(2\pi\nu t)] \times 10^8$ V/m with $\nu = 2.5$ GHz. Such field ensures that, in addition to $180^o$ stripe domains (in which dipoles are lying parallel or antiparallel to the $z$ axis inside the "up" or "down" domains, respectively), there is a spontaneous polarization lying along the $z$ axis whose magnitude can change with time but never switches in direction (it remains positive) during the entire MD simulation. Such procedure was suggested in, e.g., Ref. [28], and prevents the heating of the sample [29].

The results are analyzed by calculating the macroscopic electric polarization $\langle \mathbf{P} \rangle$ and electric toroidal moment $\mathbf{G}$ as a function of time $t$ [30]:

$$\begin{aligned} \langle \mathbf{P} \rangle &= \tfrac{1}{V} \sum \mathbf{p}_j \\ \mathbf{G} &= \tfrac{1}{2V} \sum \mathbf{r}_j \times (\mathbf{p}_j - \langle \mathbf{p} \rangle) \end{aligned} \tag{1}$$

where $\mathbf{p}_j$ is the dipole moment of the $j$-th atomic site, while $\langle \mathbf{p} \rangle$ is the dipole moment averaged over all the sites. $V$ is the volume of the supercell, and $\mathbf{r}_j$ is the vector locating the site $j$ inside the supercell. The time derivative of the electric toroidal moment is also computed since, according to Ref. [14], it is proportional to the dynamical change of the magnetization:

$$\mathbf{M} = \mathbf{M}_0 + \frac{d\mathbf{G}}{dt} \tag{2}$$

where $\mathbf{M}$ is the magnetization (in SI units) at time $t$, while $\mathbf{M}_0$ is the part of magnetization that is not related to the change of polarization [14].

Figure 1a shows the macroscopic polarization $\langle P \rangle$ along the $z$ axis as a function of time. It indicates that the polarization rather closely follows the *ac* applied electric field, thus, adopting a smooth behavior within some time ranges. On the other hand, there are some characteristic time points, at which the polarization exhibits some sudden change, which were numerically found to correspond to rapid modification in morphology and/or domain wall motions. Moreover, as this is shown in Fig. 1b, the toroidal moment (that is oriented along the $y$ axis, in the selected case, for which the "up" and "down" domains alternate

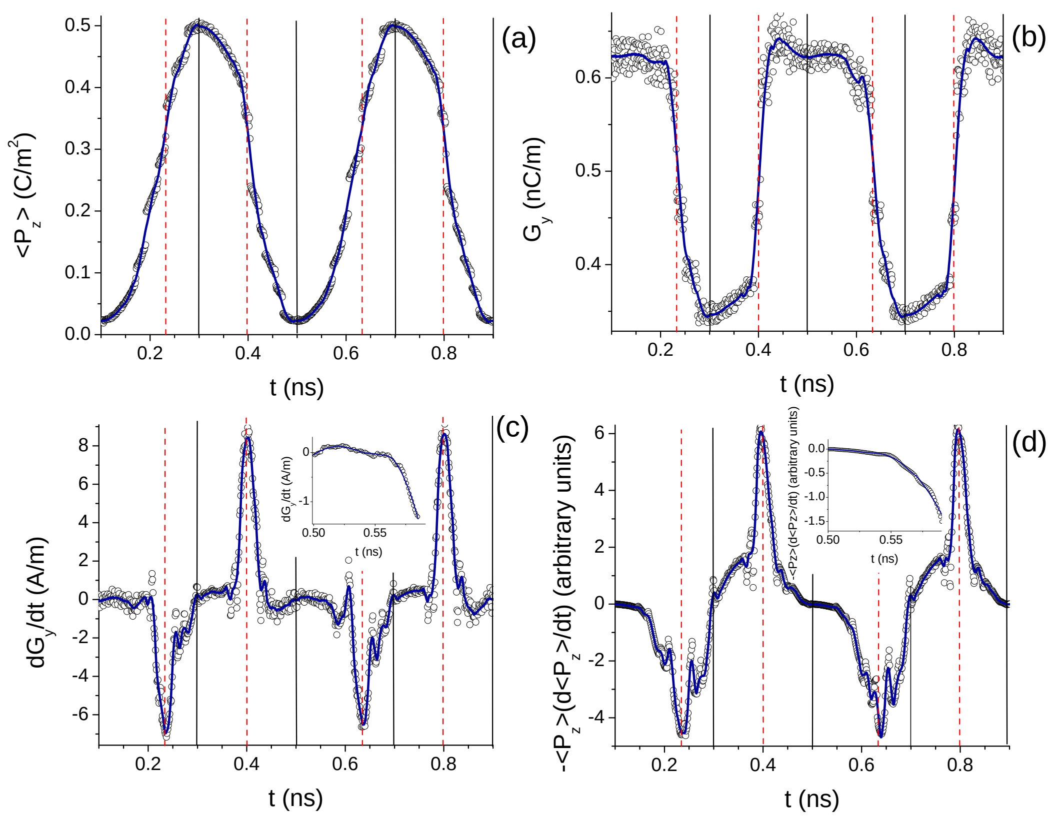


FIG. 1: (color online) Time-dependency of the electric polarization (a), electric toroidal moment (b), time derivative of the electric toroidal moment (c), and (minus) the product of the macroscopic polarization and its time derivative (d) in the studied PZT film being under the *ac* electric field described in the text. Solid vertical lines mark the position of the extrema of the polarization (which coincide with those of the electric field), while dashed lines correspond to the time associated with the pulses in the time-derivative of the electrical toroidal moment. In Panels (a) and (b), the dot symbols represent the MD data, while the solid lines are fittings of such data [31]. These fittings are then used to obtain the curves shown in Panels (c) and (d).

along the $x$ axis) exhibits large time windows inside which it varies only slightly. These small variations result in weak (but non-zero) $\frac{dG_y}{dt}$, as shown in the inset of Fig. 1c for the time interval between 0.5 and 0.6 ns. As a result, magnetization of the order of up to 1 A/m is expected to exist in this time interval, based on the direct relationship between $\mathbf{M}$ and $\frac{d\mathbf{G}}{dt}$, provided by Eq. (2). In addition, very fast variations of the toroidal moment between large ($\simeq 0.62$ nC/m) and smaller ($\simeq 0.35$ nC/m) values also occur around some specific time points. As shown in Fig. 1c, such latter fast variations automatically result in pulses for the time derivative of the toroidal moment [31]. In other words, *pulses of magnetization* are predicted to occur here, as a result of Eq. (2)! Note that this magnetization reaches values of the order $\simeq 7$ A/m, which is much smaller than the saturated spontaneous magnetization occurring in ferromagnetic Fe and Ni systems (that are of about $1.7 \times 10^6$ A/m and $0.5 \times 10^6$ A/m, respectively [32]). Note also that these pulses in $\frac{dG_y}{dt}$ (and thus of $M_y$) can be negative or positive, depending, of course, if the toroidal moment suddenly increases or decreases with time. Interestingly, panels a and c in Fig. 1 show that these pulses in the time derivative of the toroidal moment (and therefore in magnetization) do *not* occur at time intervals associated with the extrema of the polarization. For instance, a negative (respectively, positive) pulse of $\frac{dG_y}{dt}$ occurs at $\simeq 0.232$ ns (respectively, 0.4 ns), while the polarization adopts its first minimum and maximum at 0.1 and 0.3 ns, respectively.

Interestingly, we numerically found that these pulses found by our MD simulations can be understood if one assumes the following relationship between the time-derivative of the $y$ component of the toroidal moment and the product between the polarization and its time derivative:

$$\dot{G}_y = -\frac{\gamma}{P_0} \langle P_z \rangle \left\langle \dot{P}_z \right\rangle \tag{3}$$

where $\gamma$ is a coefficient and where $P_0$ is the z-component of the polarization in one of the ("up" or "down") domains under no electric field (note that the Supplemental Material [24] demonstrates that Equation (3) is valid in the simple case of a FDW moving along the $+x$ axis, with this FDW separating an "up" domain, inside which the polarization is along the $+z$ direction, from a "down" domain, inside which the polarization, with the same magnitude, is now along the $-z$ direction).

As a matter of fact, Fig. 1d reports our simulated MD data for (minus) the product of the macroscopic polarization and its time derivative. Comparing Fig. 1d with Fig. 1c does show a remarkable similarity between the time dependency of that product and $\frac{d\mathbf{G}}{dt}$, especially in the closest vicinity of the pulses, as consistent with Eq. (3) – implying that this Equation can thus be used to understand some results of Figs. 1(a-d). In particular, large pulses of $\dot{G}_y$ require the polarization to be, simultaneously, significant in magnitude *and* rapidly changed with time.

It is also important to realize that, in the hypothetical case of a FDW (of up and down domains with polarization pointing along $+z$ or $-z$, respectively) continuously moving along the $x$ axis with a velocity $v$, $\left\langle \dot{P}_z \right\rangle$ is directly proportional to $v$, as shown in the Supplemental Material [24] (note also that this Supplemental Material further shows that the time derivative of the electrical toroidal moment is directly proportional to the product between the polarization and a velocity for a more complex model). As a result, Eqs. (2) and (3) imply that such motion should generate a magnetization lying along the $y$ axis. This magnetization can naturally couple to an applied external magnetic field and, consequently, this magnetic field can facilitate or complicate the reorientation of FDW or its motion along a particular direction. As a result, Eqs. (2) and (3) provide a successful explanation for the observation of Refs. [1–4], demonstrating the existence of a magnetization in, and/or effects of applied magnetic fields on, the *moving* phase boundary and FDWs. Moreover, Ref. [4] measured a magnetic moment of $10^{-12}$ Am$^2$ in gadolinium molibdate, which corresponds to a magnetization of 0.0125 A/m for the geometry used in Ref. [4] (that is $l = 4 \times 10^{-3}$ m, $d = 4 \times 10^{-4}$ m, and $\lambda = 5 \times 10^{-5}$ m), which is about 600 times smaller than our predicted value of 7 A/m associated with the peaks of $\dot{G}_y$ seen in Fig. 1c. However, this value of 0.0125 A/m corresponds to a measured speed of the domain wall of 0.2 m/s [4] in gadolinium molibdate. Choosing now a speed of 40 m/s for the domain wall motion (which is the value recently measured for the nanosecond dynamics of ferroelectric domain walls in PZT thin films in Ref. [35]) should thus make the magnetization increasing from 0.0125 A/m to 2.5 A/m – since the magnetization is directly proportional to the velocity in our model (via the dependence of the time-derivative of the toroidal moment on velocity, see Eq. (16) of the Supplemental Material). This latter number has thus the same order of magnitude than our predicted value of 7 A/m, implying that our present predictions can be quantitatively checked in thin films experiencing nanosecond dynamics (note also that the polarization of PZT systems is larger than that of gadolinium molibdate. As a result, Eqs. (2) and (3) tell us that the magnetization of PZT thin films should even be further enhanced with respect to the estimated aforementioned value of 2.5 A/m).

Interestingly, *another simple model* detailed in the Supplemental Material implies that electric dipoles "simply" changing in magnitude with time inside *non-moving* FDWs can also create a magnetization, that is can give rise to non-zero $\frac{d\mathbf{G}}{dt}$ (see Eq. (22) of the Supplemental Material). In order to determine what are the precise *microscopic origin(s)* of $\frac{d\mathbf{G}}{dt}$, including its pulses, found in our simulations and reported in Fig. 1c, we decided to depict in Fig. 2

snapshots of the local electric dipoles in a given (x,z) plane at four different time points.

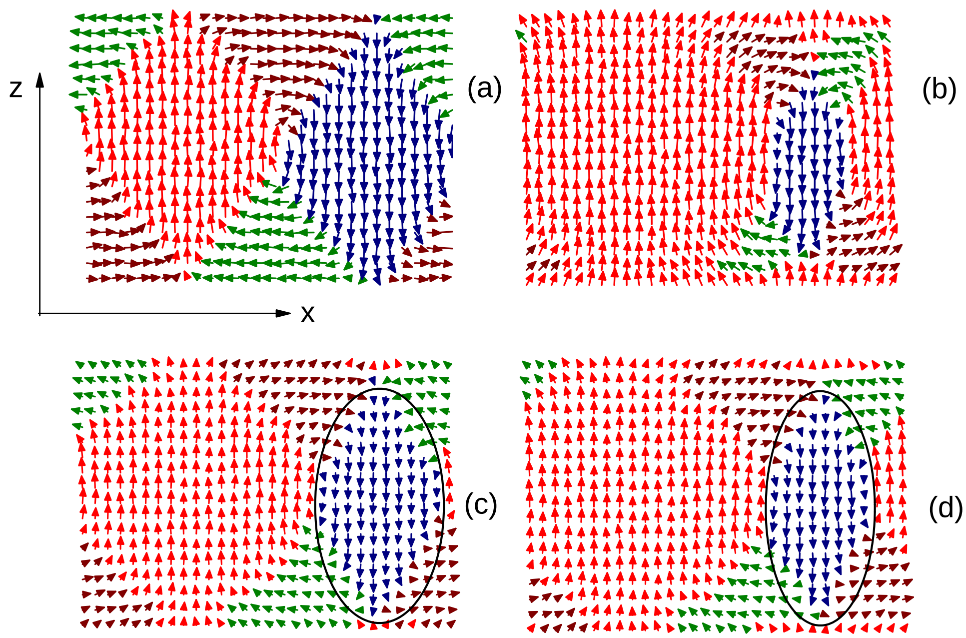


FIG. 2: (color online) Snapshots of the dipole patterns in a (x,z) plane at four different time points: (a) 0.1 ns, (b) 0.3 ns, (c) 0.230 ns, and (d) 0.232 ns. The circled areas in Panels (c) and (d) represent the area inside which the dipole pattern significantly changes within a small time variation.

Figure 2 (a) corresponds to a time at which the macroscopic polarization and applied electric fields are minima (namely, $t = 0.1$ ns). In that case, the dipolar pattern is similar to the "nominal" (i.e., under no electric field) $180^o$ stripe domains and thus also contains flux-closure domains near the film surfaces [20]. The domains with up and down polarization are, approximately, of the same width, and the FDW locates near the middle of the supercell. As shown in Fig. 2a, the flux-closure near the surfaces and the occurrence of up and down domains naturally lead to rotating dipoles inside the system, which therefore generates a significant electrical toroidal moment along the $y$ axis. Figure 2(b) corresponds to $t = 0.3$ ns, that is to a time at which the macroscopic polarization is the largest (as a result of the maximum possible value of the applied electric field). One can see that, here, the "up" domain has become much wider than the "down" domain, resulting in the motion of the FDW along the $x$ axis with respect to the first case. Note also that the flux-closure domains near the surface have shrunk, when going from Fig. 2a to Fig. 2b, which further contributes to the significant decrease of the toroidal moment between 0.1 and 0.3 ns (see Fig. 1b). We numerically found that the dipolar patterns of Fig. 2a and Fig. 2b do not evolve too much for time intervals around 0.1 ns and 0.3 ns, respectively (and for the time points derived by adding to 0.1 ns or 0.3 ns $0.4 \times n$ ns, where $n$ is an integer number), since these time intervals correspond to the extrema of the electric field and, thus, to *vanishing* time derivatives of these fields. This explains why the time derivative of the electric toroidal moment is relatively small in these time intervals (see Fig 1c and its inset). For these time points, it is, in fact, the small variation of the magnitude of electric dipoles, rather than the motion of the FDW, that is responsible for the evolution of the polarization with time and, thus, leading to the small change in $\dot{G}_y$. On the other hand, comparing Figs 2c and 2d (corresponding to $t = 0.230$ and $t = 0.232$ ns, respectively) reveals that the dipolar pattern rapidly evolves with time, for the time intervals being around those associated with the pulses of $\frac{d\mathbf{G}}{dt}$. For instance, the width of the down polarization domain abruptly shrinks from 10 to 8 lattice parameters in the middle of the down domain, between 0.230 and 0.232 ns, which, thus results in a fast increase of $\langle P_z \rangle$ – therefore explaining the negative pulse seen in $\dot{G}_y$ around 0.232 ns, according to Eq. (3). In other words, a sudden evolution of the morphology of the domain structure (via the move of the FDW) is found here to be the origin of the pulses of $\frac{d\mathbf{G}}{dt}$, and is, thus, also predicted to generate pulses of magnetization, according to Eq. (2) (relating the time derivative of the electrical toroidal moment and magnetization) [36].

Let us further indicate that we also numerically check (not shown here) how various physical parameters can affect the results reported here. For instance, we investigated a $60 \times 4 \times 20$ supercell, i.e. that is twice larger along the x-axis than the one studied in the manuscript. Such elongation results in the occurrence of four (rather than two) alternating up and down domains within this supercell, but has merely no effect on the results shown in Fig. 1. Similarly, we also find *qualitatively* similar results to those shown in Fig. 1, including the occurrence of pulses in the time derivative of the electrical toroidal moment, when: (i) investigating a $20 \times 4 \times 8$ supercell, which corresponds to a film's thickness (along the z-axis) being 2.5 times smaller than the one investigated here, with this film being the subject of the electric field $E(t) = [3.0 - 2.0 \times sin(2\pi\nu t)] \times 10^7$ V/m, where $\nu = 2.5$ GHz; and (ii) changing the frequency of the applied *ac* field from 2.5 GHz to 5.0 GHz, for our studied $30 \times 4 \times 20$ supercell. The magnitude of the pulses of the time derivative of the electrical toroidal moment (and thus of the pulses in magnetization) does depend on these parameters. For instance, increasing the field frequency enhances the magnitude of these pulses, while decreasing the thickness of the film, and, simultaneously, decreasing the strength of the electric field (in order to avoid full poling of the sample), reduces the magnitude of these pulses.

Let us now compare our results with previously proposed theories related to ME effect associated with dynamics of FDW [4, 5]. First of all, one can note that Eqs. (2) and (3) do not require the motion of a polarized medium existing between the up and down domains to explain such results, in contrast to Ref. [5], and that such polarized medium is not found in our present simulations of ultra thin films. Regarding the fundamental assumption of Ref. [4] about a closed loop of the electric current density located near the FDW, we also found it to be invalid in our studied systems. As revealed in the Supplemental Material [24], such current density rather mostly posseses lines oriented along the $+z$ (respectively, $-z$) direction and located near the FDW, for the time intervals corresponding to a negative (respectively, positive) pulse of the electric toroidal moment.

In summary, this computational work predicts the existence of pulses of magnetization when PZT ultra thin films undergo sudden changes of morphology of their nanodomain structure, as a response to an *ac* electric field. Presently-developed simple models can explain not only the occurrence of such pulses, but also puzzling dynamical magnetoelectric effects that have been reported about 30 years ago [1–3]. We also numerically determine that electric dipoles changing in time within non-moving domains can generate a magnetization too, but of much weaker magnitude, and develop another simple model demonstrating that fact. We are thus confident that the present results deepen the current knowledge of nano science, domain walls, ultrafast dynamics, ME effects and electromagnetism [37]. Our presently discovered effect of the occurrence of magnetization pulses may lead to the design of original devices exploiting such striking dynamical ME effect.

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[24] The Supplemental Materials (1) provide details about our numerical method ; (2) also derive Eq. (3) from some simple models about *moving* ferroelectric domains; (3) further demonstrate that magnetization can also be created when electric dipoles "only" change their magnitudes within *non-moving* domains; and (4) report the computation of the electric current density in our case.

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[31] Note that the MD data shown in Figs 1a and 1b are smoothed [33] with the use of polynomials of the second order to extract the time derivatives of the polarization and toroidal moment.

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[36] Interestingly, we performed additional analysis for which the "horizontal" parts of the flux-closure structure seen in Fig. 2 were excluded in the computation of the toroidal moment. The resulting toroidal moment-versus-time function was basically found to be identical to the one shown in Fig. 1b (because the horizontal parts of the flux closure of different neighboring domains seen in Fig. 2 cancel each other in this computation). Such fact naturally implies that our results are general in nature (i.e. they are not only valid for flux-closed structures)

and also explains why the simple $180^0$-domain-wall model documented in the Supplementary material can reproduce well our numerical results.

[37] Let us note that the time derivative of the polarization can generate a curl of magnetization according to laws of electromagnetism. Interestingly, in case of a homogeneous polarization, one can show that the macroscopic average of such magnetization will vanish. On the other hand and as shown in Ref. [14], a non-zero macroscopic magnetization can be created by a non-zero time-derivative of the electrical toroidal moment (which is precisely the case for the studied domain structure under ac electric field and which is associated with inhomogeneous polarization field).