

# Origin of jump-like dynamics of the plane domain wall in ferroelectrics

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In this paper, we demonstrate how the experimentally observed nonmonotonic sideways motion of the plane domain wall in uniaxial ferroelectric, caused by the electric circuit feedback, can be described theoretically on the basis of the kinetic approach. We demonstrated experimentally the appearance of self-sustained periodic oscillations of the domain wall velocity and compared the theoretical results with the experimental data for switching in model ferroelectric stoichiometric lithium tantalate ( $\text{LiTaO}_3$ ) produced by vapor transport equilibration (VTE-LT). Quasiperiodic and chaotic regimes of wall motion are predicted to be observed experimentally.

**Keywords:** sideways wall motion, kinetic approach, retardation of screening, nonlinear oscillator, lithium tantalate, quasiperiodic oscillations of wall velocity

**Short title:** Jump-like dynamics of the plane domain wall

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## 1. Introduction

The polarization reversal, being an attribute of ferroelectrics, has been always studied intensively. It has been shown that domain kinetics obtained in various ferroelectric materials essentially depends on experimental conditions. The understanding of the domain evolution mechanisms is necessary for creation of the precise tailored domain structures [1-3]. This technology denoted as “domain engineering” is developing rapidly [4, 5]. The periodically poled nonlinear optical materials are widely used for the production of the tunable coherent light sources based on quasi-phase matching effect [6-10].

At present, the thermodynamic approach is applied usually for analysis of the polarization reversal process and domain kinetics in ferroelectrics [11]. Though a large amount of experimental data concerning the domain structure evolution is accumulated, its theoretical description is developed to a much less degree. Our current understanding of the domain structure evolution is based on the kinetic approach, which takes into account the analogy of the domain kinetics in electric field to the first-order phase transition achieved through nucleation [3, 12, 13]. All stages of the domain kinetics have been attributed to the elementary processes of the thermally activated nucleation leading to growth of domains with preferred orientation of the spontaneous polarization defined by direction of the applied field. The nucleation probability is always spatially heterogeneous, as it is determined by the local value of the electric field produced by several sources [3, 12].

The kinetic approach is flexible enough to analyze a multitude of experimentally observed situations. In particular, it allows one to explain the origin of the variety of experimentally observed domain shapes, to predict qualitatively different regimes of the domain wall (DW) motion and to evaluate the critical values of the relevant parameters, which define transitions between these regimes [14-23]. However, there is no straightforward and general way at hand to extract the quantitative information about the process from the

experimental data.

Sideways DW motion under application of the electric field is the stage of the domain kinetics studied experimentally by *in situ* methods in the best way. In some model crystals it is possible to obtain the motion of the plane DW with constant shape [15, 16, 18-20]. Several variants of this simplest process have been revealed. It has been demonstrated that the wall motion can be essentially nonmonotonic, such as the jump-like (jerky) wall motion in congruent lithium niobate  $\text{LiNbO}_3$  (LN) (Fig. 1) [15, 16] and gadolinium molybdate  $\text{Gd}_2(\text{MoO}_4)$  (GMO) single crystals [18, 19, 22, 23]. The process has been attributed to the deceleration of DW motion due to incomplete bulk screening and to interaction of the moving DW with the bulk defects [15-19, 22, 23]. Moreover, it has been demonstrated that the nonmonotonic DW motion can be realized also during polarization reversal in the circuit with the series resistance. In this case, the type of DW dynamics is defined by experimentally controlled feedback and delay.

In this paper, we demonstrate how the nonmonotonic scenario of DW motion caused by the circuit feedback can be described theoretically on the basis of the kinetic approach [3, 12]. Actually, in the framework of the kinetic approach one assumes that the local value of the electric field, being composed of several components with different physical origins, is the driving force of the nucleation process:

$$\vec{E}(x,t) = \vec{E}_{ext} + \vec{E}_{dep}(x,t) + \vec{E}_{ext.sc}(x,t) + \vec{E}_{int.sc}(x,t) \quad (1)$$

where the terms are external, depolarization, external screening, and internal screening fields, respectively.

Eq. (1) is supplemented by phenomenological equation of the field dependence of DW motion velocity ( $v = v(E)$ ). The latter may have the following form, which corresponds to experimental data obtained in LN and GMO crystals [15, 16, 18, 19]:

$$v(E(t)) = \mu (E_{loc}(v(t)) - E_{th}), \text{ for } E_{loc} > E_{th} \quad (2)$$

$$v(E(t)) = 0, \text{ for } E_{loc} < E_{th},$$

where  $E_{th}$  is the threshold field,  $\mu$  is the DW mobility.

One should mention that in literature (especially in the theoretical one) there is a tendency to solve the system of Eqs. (1) and (2) as if it has the only solution with constant velocity. In particular, such assumption was made by Morozovska et al. [11], where the influence of the screening retardation on the motion of the plane DW was taken into account. The authors established the existence of a stable steady DW motion for low and high velocity. However, they discovered the appearance of the DW motion instability in the range of intermediate velocities. Naturally, the actual type of the unsteady DW dynamics could not be determined within their simplified approach.

The difficulty of the correct solution of the system (1), (2) is caused by the complex dependence of the field components in Eq. (1) on the unknown DW path of motion. Namely, the local field value is dependent not only on the current DW coordinate  $X(t)$ , but on its previous positions  $X(t' < t)$ , also. Therefore, the simple form of the Eq. (1) is deceptive. Furthermore, the crucial dependence of the DW motion scenario on the memory effects caused by the screening retardation was confirmed by numerous experiments [13-23].

Of course, one can ignore the analytical difficulties and try to solve the system (1), (2) numerically using reasonable assumptions about the possible DW dynamics by analysis of the series of momentary domain patterns obtained by *in situ* visualization of the domain structure evolution during polarization reversal [3, 12]. However, our understanding of the fundamental switching mechanisms would be much deeper and the whole description of the processes would reach much higher quantitative level, if instead of a rather symbolic form (1), (2) the equation of DW motion assumed the explicit form:

$$F(X(t), \dot{X}(t), \ddot{X}(t), \ddot{\ddot{X}}(t), \dots) = E_{ext}(X(t), t) \quad (3)$$

where  $X(t)$  is the position of the DW.

It would be especially fine, if it turned out to be possible to reduce approximately the Eq. (3) to such a form that is simple enough to obtain the explicit solutions in an analytical way, which would be the valuable step in the development of the kinetic approach.

The goal of the present paper is twofold. We consider the switching process in ferroelectric capacitor, which is accomplished by single DW motion. In particular, we derive the DW motion equation, which turns out to be of the type of Eq. (3). Namely, it takes the form of a nonlinear oscillator equation that has “a negative friction force” within a certain electric field range. We establish the appearance of self-sustained periodic oscillations of the DW in this range and compare our theoretical results with the experimental data for switching in model crystal stoichiometric lithium tantalate ( $\text{LiTaO}_3$ ) produced by vapor transport equilibration (VTE-LT) [16, 24].

## **2. Experimental observation of nonmonotonic DW motion in VTE-LT**

Switching current was recorded during polarization reversal in VTE-LT single crystal in the circuit with series resistance ( $R = 10 \text{ M}\Omega$ ) by application of the rectangular electric field pulses with amplitude 300-500 V (Fig. 1).

The studied samples represented 800- $\mu\text{m}$ -thick plates cut normal to the polar axis with both polar surfaces covered by liquid electrodes (water solution of LiCl). The typical shape of the switching current (Fig. 2) contains the number of the narrow peaks corresponding to short-time accelerations of the wall motion. The amplitude and average frequency of the current peaks depend on the circuit parameters.

## **3. Electric circuit and capacitor geometry**

Let us consider an electric circuit containing voltage source  $U_0$ , ferroelectric capacitor  $C$ , and series resistance  $R$ . The voltage  $U(t)$  applied to the ferroelectric capacitor which

depends on the voltage drop on the series resistance is given by:

$$U(t) = U_0 - I(t)R \quad (4)$$

where  $U_0$  is the voltage applied to the circuit,  $I(t)$  is the switching current,

$$I(t) = C \frac{dU(t)}{dt} - 2P_s v(t) l_{DW}, \quad (5)$$

where  $P_s$  is the spontaneous polarization,  $l_{DW}$  is the DW length,  $v(t)$  is the sideways DW velocity.

It is seen that the presence of the series resistance provides the negative feedback, which stabilizes the circuit current near its average value. However, the real time dependence of switching current can be nontrivial due to the existence of the ferroelectric capacitor, which plays the role of a nonlinear element with memory, as it will be discussed in detail later on.

The schematic representation of the ferroelectric capacitor with the intrinsic or artificial surface dead layer (dielectric gap) located at the interface between ferroelectric plate and electrode [11] is shown in Fig. 3.

The studied ferroelectric capacitor represents the plate of uniaxial ferroelectric crystal with thickness  $d$  cut normal to polar axis with polar surfaces covered by continuous electrodes. The intrinsic or artificial dielectric gap (dead layer) with thickness  $L$  is situated between the electrode and the surface of the ferroelectric plate (Fig. 3). The screening charge layer with thickness  $h_{sc}$  is situated in ferroelectric under the dead layer. The studied neutral 180-degree DW represents a single vertical plane moved along  $X$  direction under the action of the electric field exceeded the threshold value  $E_{th}$ .

#### 4. Screening retardation and electric field on the nucleus

We will assume that the polarization profile of the moving DW doesn't change and can be approximated by  $P_0(x - X(t))$ , where  $X(t)$  is the current DW coordinate. The residual depolarization field behind the DW is screened partially by the charge localized at the

interface with density  $\sigma(x,t)$ . In the case when the screening mechanism does not involve any long-range diffusion of the carriers (for instance, if the screening is caused by the redistribution of the bulk charges or by reorientation of the defect dipoles) [3, 12], the corresponding phenomenological relaxation equation for  $\sigma(x,t)$  can be written in the following form [11]:

$$\tau \frac{d\sigma(x,t)}{dt} = P_0(x - X(t)) - \sigma(x,t) \quad (6)$$

where  $\tau$  is the characteristic time of the screening process.

The solution of the equation is given by:

$$\sigma(x,t) = \frac{1}{\tau} \int_{-\infty}^{\infty} P_0(x,t') e^{\frac{t-t'}{\tau}} \theta(t-t') dt' \quad (7)$$

where  $\theta$  is Heaviside function.

We assume that the DW kinetics is determined by the nucleation at the wall near the polar surface, where the local electric field  $E$  is maximal. Correspondingly, one needs to calculate the value  $E(z=0, x=X(t), t)$ , which is given by:

$$E(z=0, x=X(t), t) = - \int_{-\infty}^{\infty} \frac{e^{-ikX(t)}}{\sqrt{2\pi}} \tilde{\sigma}_{eff}(k,t) f(k,L) dk \quad (8)$$

where  $\tilde{\sigma}_{eff}(k,t) = \tilde{P}_0(k,t) - \tilde{\sigma}(k,t)$  is the Fourier transform of the density of net charge and the function  $f(k,L)$  is defined by:

$$f(k,L) = \frac{\tanh(Lk) \cosh\left(\frac{k(d-L)}{\gamma}\right)}{\varepsilon_0 \left( \varepsilon_3 \tanh(Lk) \cosh\left(\frac{k(d-L)}{\gamma}\right) + \gamma \varepsilon \sinh\left(\frac{k(d-L)}{\gamma}\right) \right)} \quad (9)$$

where  $\gamma = \sqrt{\frac{\varepsilon_3}{\varepsilon_1}}$  is anisotropy factor,  $\varepsilon_1, \varepsilon_3$  are permittivity for XY and Z directions,  $\varepsilon$  is permittivity of dead layer,  $\varepsilon_0$  is the electric constant.

Combining the exponential factor in Eq. (8) with a similar one taken from the Fourier transform  $\tilde{\sigma}_{eff}(k,t)$ :

$$\tilde{\sigma}(k,t) = \frac{1}{\tau} \int \tilde{P}_0(k,t') e^{ikX(t')} e^{\frac{t-t'}{\tau}} dt' \quad (10)$$

one can expand their combination coming into the integrand (8):

$$e^{ik(X(t')-X(t))} \approx e^{ikv(t)} \left[ 1 + \frac{ik}{2} \dot{X}(t)(t'-t)^2 + \dots \right] \quad (11)$$

Now we make the assumption that one can truncate the series in Eq. (11) neglecting all higher order acceleration contributions. It is clear that the assumption is trivially correct in case of the steady-state DW propagation regime. However, we will prove that it can be a good approximation in the unsteady DW regime as well. Under this assumption, the integration over time in Eq. (8) can be performed as:

$$\int_{-\infty}^{\infty} e^{ik(X(t')-X(t))} e^{\frac{t'-t}{\tau}} \theta(t-t') dt' \approx -\frac{i\tau(k\dot{X}(t)\tau-i)^2 + k\tau^3\ddot{X}(t)}{(k\dot{X}(t)\tau-i)^3} \quad (12)$$

Accordingly, the electric field on the nucleus consists of two contributions, which depend on the DW velocity  $\dot{X}(t)$  and acceleration  $\ddot{X}(t)$ , respectively. Making use of Eq. (12) the next integration over  $k$  can be done in Eq. (8) in the limit  $d \gg L$  and  $kL \ll 1$ .

Taking into account the additional electric field contribution coming due to a negative feedback  $E_{nf}$  (see Eq. (4)) under the assumption that  $\tau \ll \tau_{ec} = RC$  one finds:

$$E_{nf}(h(t)) = \frac{U_0}{d} - \frac{2lP_sRh(t)}{d\tau_{ec}} \quad (13)$$

where  $h(t) = X(t) - v_0t$  and  $v_0$  is the average DW velocity, which is defined in the experiments by series resistance  $R$ .

Substituting the result and also the delayed electric field contribution into Eq. (2), one arrives at the following equation of the DW motion:

$$M(\dot{X}(t)) \ddot{h}(t) + F(\dot{X}(t)) - E_{th} + \frac{2lP_sRh(t)}{d\tau_{ec}} - \frac{\dot{X}(t)}{\mu} = 0 \quad (14)$$

where  $M$  is the effective mass of the DW.

Notice, that the Eq. (14) just has the form described in the introduction. The velocity dependence of  $M(V)$  is represented in Fig. 4.

The following parameters have been used for numerical simulation:  $P_s = 65 \mu\text{C}/\text{cm}^2$ ,  $d = 0.8 \text{ mm}$ ,  $U_0 = 400 \text{ V}$ ,  $E_{th} = 50 \text{ V}/\text{mm}$ ,  $\varepsilon = 20$ ,  $\varepsilon_l = 80$ ,  $\varepsilon_3 = 30$ ,  $\tau = 1 \text{ s}$ ,



$\mu = 20 \text{ mm}^2/(\text{kV}\cdot\text{s})$ ,  $l_{DW} = 2 \text{ mm}$ ,  $R = 10 \text{ M}\Omega$ ,  $C = 3.5 \text{ pF}$ .

## 5. Solutions of the DW motion equation

The dynamics of the forced nonlinear oscillator is described by Eq. (14). In case of the application of the constant external electric field (infinite rectangular pulse) it is convenient to define  $\dot{X}(t) = v_0 + \dot{h}(t)$ , where  $v_0$  is the average DW velocity. The value of  $v_0$  is defined by the following algebraic equation:

$$\frac{v_0}{\mu} = \frac{U_0}{d} - E_{th} - F(v_0) \quad (15)$$

One has the nonlinear differential equation to find the time-dependent function  $h(t)$ , which describes the deviation from the steadily propagating DW:

$$M(v_0 + \dot{h}(t))\ddot{h}(t) + \frac{2lP_sR}{\tau_{ec}d}h(t) + \tilde{F}(v_0 + \dot{h}(t)) = 0 \quad (16)$$

where  $\tilde{F}(\dot{X}(t)) \equiv F(\dot{X}(t)) - F(v_0) + \frac{\dot{X}(t)-v_0}{\mu}$ .

If the derivative  $\frac{d\tilde{F}(\dot{X})}{d\dot{X}}$  taken at  $\dot{X}(t)=v_0$  is positive, then steady-state solution (with  $h = 0$ ) is stable. According to Eq. (15),  $F(v) + \frac{v}{\mu} = E_0 - E_{th}$  plays the role of friction force. However, as it is seen from Fig. 5, the range of the average velocities (within the corresponding interval of the external electric field  $E_0 = \frac{U_0}{d}$ ) can show the point, where the derivative becomes negative. Then the friction force term in Eq. (16) is also negative, which means physically that there is energy input to the DW motion. The input provides the appearance of the stationary self-sustained oscillations of  $h(t)$  and the corresponding periodic oscillations of the DW velocity around its average value  $v_0$ . The characteristic feature of the oscillations is that they typically deviate strongly from the conventional harmonic ones, except the value  $v_0$  turns out to be very close to the end points of the instability interval ( $\frac{d\tilde{F}(\dot{X})}{d\dot{X}} = 0$ ). The switching current  $I(t)$  is presented in Fig. 6.

Now we can at last justify our main assumption that the higher order acceleration contributions in Eq. (11) do play a minor role. Looking at Fig. 6, one notices that within the most part of the period the DW moves almost steadily with small acceleration. The short fractions of period, where the DW accelerates and decelerates drastically, are potentially dangerous for the truncation of Eq. (11). It means that the truncation procedure is good everywhere except inside the “fine structure” of the narrow current peaks. We can also confirm the visual picture by the analytical argument. If one scales both the time  $t$  and spatial coordinate  $x$  by the same factor  $w \equiv \frac{\tau_{ec} d}{2lP_s R}$ , Eq. (16) assumes the following form:

$$M(v_0 + \dot{h}(t))w\ddot{h}(t) + w\tilde{F}(v_0 + \dot{h}(t)) + h(t) = 0 \quad (17)$$

The mass term is multiplied by the factor  $w$ , while the velocity value is conserved under the scaling transformation. In case when the factor is small, it is clear that each additional differentiation over time in the higher order accelerations would be accompanied by the higher power of the same small factor. Therefore, the weak feedback regime can be quantitatively described by the truncated Eqs. (15) and (16).

Eq. (17) is so called generalized Rayleigh equation. It is known that in case  $\frac{d\tilde{F}(\dot{X})}{d\dot{X}} > 0$  it has a stable fixed point  $h = 0$ ,  $dh/dt = 0$ , which corresponds to a steady state propagation of DW with the constant velocity  $v = v_0$ . In the opposite case  $\frac{d\tilde{F}(\dot{X})}{d\dot{X}} < 0$  a stable limit cycle appears, which is drawn in Fig. 7.

In the unsteady regime, the propagation of DW is accompanied by the oscillations of velocity around its average value  $v_0$ . After finishing of the transient stage, the DW motion assumes the perfect oscillatory character shown in Fig. 8.

The irregular current oscillations are seen in experimental results (Fig. 2). This fact can be attributed to presence of immobile “quenched” defects [17]. To describe their role one should include the corresponding stochastic term in Eq. (16).

Similar equations have been explored in a general context of nonlinear dynamics, and a lot of information about the properties of their solutions has been obtained already [25]. For instance, if the applied field pulse is more complicated than the rectangular one, the straightforward extension of Eq. (16) assumes the form of so called perturbed Rayleigh equation [26]. Even in the simplest case of a single DW the latter non-autonomous equation has 3D phase space. Then, in contrast to the time-independent (“infinite”) rectangular pulse of the electric field, the structure of the attractor can be much more complicated than simple fixed points and limit cycles [26].

## **6. Conclusion**

In the paper, we have demonstrated how the general concepts of the kinetic approach can be further developed to obtain the quantitative description of the interplay between the domain kinetics and screening retardation. In particular, it has been shown that under certain conditions the memory effects caused by the screening processes can be taken into account using nonlinear differential equations. Similar equations have been investigated in a general context of nonlinear dynamics, and a lot of information about the properties of their solutions has been obtained already. Such phenomena as period doubling, synchronization, quasiperiodicity, and chaotic regimes are predicted to be observed in the measured current. The outlined analytical method can be used also to consider more complicated switching problems (like a growth of isolated polygon-shape domain, etc.). We are going to study them in future publications.

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## Figure captions

Fig. 1. Scheme of the electric circuit with ferroelectric capacitor and series resistance.

Fig. 2. Switching current measured for polarization reversal of VTE-LT crystal in the circuit with series resistance ( $R = 10 \text{ M}\Omega$ ).

Fig. 3. The scheme of the ferroelectric capacitor with dielectric gap [11].

Fig. 4. The calculated dependences of the DW effective mass  $M(v)$  on velocity for various thicknesses  $L$  of the dielectric gap:  $L = 100 \text{ nm}$  (blue thick curve),  $L = 1 \text{ }\mu\text{m}$  (red dashed curve),  $L = 10 \text{ }\mu\text{m}$  (green dot-dashed curve).

Fig. 5. The calculated dependences of the DW velocity  $v$  on friction force for various thicknesses  $L$  of the dead layer according to Eq. (15).

Fig. 6. The time dependence of the switching current for thickness of the dead layer  $L = 0.1 \text{ }\mu\text{m}$ , average value of velocity  $v_0 = 5.85 \text{ mm/s}$  and with initial conditions  $h(0) = 0$ ,  $h'(0) = 40 \text{ mm/s}$ .

Fig. 7. Incipient limit cycle for thickness of the dead layer  $L = 0.1 \text{ }\mu\text{m}$ , average value of velocity  $v_0 = 5.85 \text{ mm/s}$  and with initial conditions  $h(0) = 0$ ,  $h'(0) = 1 \text{ mm/s}$ .

Fig. 8. Transferring of transient regime to limit cycle, for thickness of the dead layer  $L = 0.1 \text{ }\mu\text{m}$ , average value of velocity  $v_0 = 5.85 \text{ mm/s}$  and with initial conditions  $h(0) = 0$ ,  $h'(0) = 1 \text{ mm/s}$ .