Pis'ma v ZhETF, vol. 119, iss. 10, pp. 766 – 767

Impact of domain wall conduction on ferroelectric domain reversal kinetics[1)]

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Submitted 14 March 2024
Resubmitted 15 April 2024
Accepted 16 April 2024

DOI: 10.31857/S1234567824100100, EDN: VWBMGV

Ferroelectric polarization reversal is a vast developing research area [1]. Reversal of the spontaneous polarization P occurs at electric fields E much smaller than the depolarization field $E_d = 4\pi P/\varepsilon_c$ via nucleation of small critical domains and their subsequent fast-forward and lateral growth. The classical models for the domain formation energy [1–3] deal with the dielectric case (no domain wall (DW) conduction). They lead to non-realistically high energies and are not consistent with the most common law $t_R \propto \exp(E_*/E)$ for the reversal time. The exponential $\exp(-E_l/E)$ Merz law [4] for the lateral expansion with $E_l < E_*$ is proven for many materials including lithium niobate (LN) crystals [1].

Recently, DW conduction was discovered and explored in many materials [5, 6] to become a general ferroelectric effect. It can also be regarded as a general mechanism for the charge compensation during the reversal. The absence of this mechanism would lead to huge depolarizing fields and contradictions within any compensation-free polarization reversal concept. The impact of DW conduction on the polarization reversal remains unexplored. It is indicated, however, that DW conduction substantially modifies the classical expressions for the domain formation energy [7, 8].

We show that account for the DW conduction in LN crystals leads to the exponential $\exp(-E_{n,l}/E)$ field dependences for the rates of domain nucleation (n) and lateral (l) growth with characteristic fields $E_{n,l}$ comparable with E_* and the ratio $E_n/E_l \approx 5$ controlled by the crystal symmetry. Modeling of the reversal kinetics shows distinct stages of nucleation, lateral growth, and coalescence. The field dependence of the reversal time obeys, in accordance with experiment [9], the law $t_R \propto \exp(E_*/E)$ with $E_* \approx 35\,\mathrm{kV/mm}$.

An important ingredient of our theory is the assertion that the DW conduction not only lowers the critical domain formation energy W_*, but also provides the necessary E_n/E dependence on the applied electric field. This result was missed in [7, 8]. The corresponding relation for W_* for a semi-spheroidal nucleus is

$$W_* \simeq \pi l_a^{*2} w_1 + \frac{4\pi w_0 l_a^* l_c^*}{3}; \quad l_c^* = \frac{\sqrt{4\pi\Lambda w_0 l_a^*}}{E\sqrt{\varepsilon_a}}, \qquad (1)$$

where the critical transverse size $2l_a^*$ is about 1 nm, $\Lambda = \ln(2l_c^*\sqrt{\varepsilon_a}/l_a^*\sqrt{\varepsilon_c})$, $w_{0,1}$ are the surface energies for neutral (0) and maximally charged (1) sections of DW, and $\varepsilon_{a,c}$ are the dielectric constants. This relation provides the inequality $l_c^* \gg l_a^*$ for typical values of E.

Also, we have shown that the presence of DW conduction leaves unchanged the result of [3] about the E_l/E law for the lateral domain expansion in the dielectric case. The characteristic fields E_n and E_l controlling nucleation of domains and their subsequent lateral growth are linked with each other by a simple relation accounting for the point symmetry. For the 3m symmetry of LN crystals $E_n/E_l = 3\sqrt{3}$.

The indicated results enable us to formulate a simple kinetic model of the domain reversal: The input surface area consists of equilateral triangle unit cells with side $a \approx 0.5$ nm. Each cell can be in two states, relevant to non-inverted and inverted domains. An inverted cell cannot flip back. Two probabilities for a cell to flip in a unit time are important. First, it is the probability for an isolated cell to flip ν_n. Second, it is the probability to flip for a close neighbor ν_l, i.e. for a cell having one common length element with already inverted one. Correspondingly, we set

$$\nu_{n,l} = \tau_*^{-1}\exp(-E_{n,l}/E), \quad E < E_l = E_n/3\sqrt{3}, \quad (2)$$

[1)]Supplementary materials are available for this article at DOI: 10.1134/S002136402460071X and are accessible for authorized users.

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where τ_* can be regarded as a free parameter with values lying in the range $(10^{-1} - 10^{-3})$ s.

Consider some simple features of the reversal kinetics within our model. Let N be the total number of unit cells to flip, $\lg N \gg 1$. The number of nucleated isolated cells grows initially as $N_n = N\nu_n t$. As soon as the rate $3N_n\nu_l$ exceeds $N\nu_n$, the lateral nucleation events become increasingly important. This leads to the formation of domain clusters and, ultimately, to domain coalescence and complete reversal.

We simulated the domain reversal kinetics for $N = 10^6$, 10^8, and 10^{10}. Each iteration $\mu = 1, 2, \ldots$ corresponds to a single flip and to a certain time step δt_μ. After each iteration, all cells are divided into four groups, A, B, C, D. The group A consists of inverted cells. The group B includes non-inverted cells possessing one common length element with the inverted ones. The group C consists of non-inverted cells possessing 2 and 3 common elements with the inverted ones. The group D consists of the non-inverted cells. The cells are renumerated in each group after each iteration.

Consider an arbitrary iteration. The time step is here $\delta t_\mu = (\nu_n N_D + \nu_l N_B)^{-1}$. Next, we introduce two probabilities $p_n = \nu_n N_D \delta t_\mu$ and $p_l = \nu_l N_B \delta t_\mu$ such that $p_n + p_l = 1$. After that, we choose randomly between p_n and p_l. With the actual process determined, we choose randomly one of the cells within the actual group.

Computer simulations were performed for $E_n = 75$, $E_l = E_n/3\sqrt{3} \simeq 14.5$ kV/mm, and $\tau_* = 0.01$ s within the range $E = (3 - 12)$ kV/mm.

Sub-figures 1a, b, c show representative domain patterns at different stages of domain reversal for $N = 10^8$ and $E = 4$ kV/cm. At 19 s we are within the stage of lateral growth. The predominating shapes of domains are close to circles. At 33 s, when about 40 % of cells are inverted, we see a broad distribution of domain forms and sizes; this is a developed coalescence stage. At $t = 50$ s, when about 90 % of cells are inverted, we can see only rare peculiar islands of non-inverted domains.

One of the most important results is the field dependence of the reversal time t_R. With a good accuracy, it follows the law $t_R = (\nu_n \nu_l^2)^{-1/3} = \tau_* \exp(E_*/E)$ with $E_* = (E_n + 2E_l)/3 \simeq 34.4$ kV/cm. This is in a nice agreement with experiment [9]. Among other findings is a scaling invariance of the dependence $N_A(t)$ for different fields and the influence of E on the fractions of domains inverted via primary nucleations and the lateral expansion.

In conclusion, it is shown that conduction of DWs in LN crystals leads to strong consequences for the field induced domain reversal: The activation energy for nucleation lowers, and the reversal kinetic represents nucleation of needle-like domains and their subsequent lateral growth resulting in the law $\exp(E_*/E)$ for the reversal time. The kinetic processes running at different fields are self-similar.

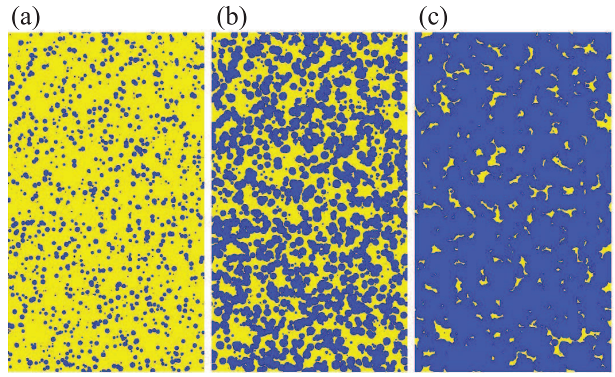

Fig. 1. (Color online) Domain patterns for $N = 10^8$ and $E = 4$ kV/mm. The cases (a), (b), (c) correspond to $t = 19$, 33, 50 s. Blue and yellow areas refer to inverted and non-inverted domains. The horizontal size is $10^4 a$

Funding. N. Masnev was supported by the Ministry of Science and Higher Education of the Russian Federation (state assignment # FFWR-2024-0014).

Conflict of interest. The authors of this work declare that they have no conflicts of interest.

This is an excerpt of the article "Impact of Domain Wall Conduction on Ferroelectric Domain Reversal Kinetics." Full text of the paper is published in JETP Letters journal. DOI: 10.1134/S002136402460071X

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