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Domain Wall Motion for Slowly Varying Electric Field

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The problem of domain wall motion near a ferroelectric/dielectric interface is studied using the Landau-Ginzburg-Devonshire three-dimensional phenomenological approach. To account for the effect of the "dead layer", the generalized boundary conditions are introduced. The small value of the dielectric permittivity in comparison with the ferroelectric one allows us to consider both half-spaces independently. The dependence of 180o domain wall velocity on the applied electric field is investigated. It is assumed that characteristic time and length of the varying external field are significantly greater than the ferroelectric relaxation time and length, respectively. In our investigations the axially symmetric ferroelectric is considered.

Keywords Ferroelectric domain; domain wall; dead layer; electrostatic field; SPM tip

I. Introduction

The application of the ferroelectric materials for memory devices is based on their ability to change the direction of the polarization as a function of the applied field. For many years this switching phenomenon in ferroelectrics has been widely studied both theoretically and experimentally [1–9]. At the present moment, one may conclude that in accordance with experimental data there exist five main stages of the ferroelectric domain structure kinetics. Namely, there are: (i) nucleation of elongated domains with inverse polarization, (ii) forward growth, (iii) sideways growth, (iv) coalescence, and (v) spontaneous back-switching. The general theory, which is able to describe all aforementioned stages, is not presented even nowadays. At the same time, the lateral movement of the domain wall is the most investigated among them. The majority of studies were carried out considering a uniform electric field. However, the necessity to take into account the non-homogeneously distributed external electric field was demonstrated in Ref. [7]. In this work we examine the case of a weak external field dependence on the time and space. Namely, it is assumed that the characteristic time and length of the varying external field are significantly greater than the relaxation time and the length of the ferroelectric material. In our consideration ferroelectric is free from defects. The main object of the study is the dependence of the domain wall motion velocity on an external electric field. The treatment is within the framework

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of the time-domain Landau-Ginzburg-Devonshire (LGD) theory. The axially symmetric ferroelectric is considered.

II. Statement of the Problem and its Simplification

Choice of the Equation

One of the standard ways to describe the electric field in ferroelectrics is the LGD model [10]. According to this approach, the free energy density $F(\eta, E)$ is written as:

$$F(\eta, E) = \frac{a\eta^2}{2} + \frac{b\eta^4}{4} + \frac{g(\nabla\eta)^2}{2} - \vec{E}\eta, \tag{1}$$

where η is the order parameter, $\vec{E}$ electric field. LGD coefficients $\{a < 0; b, g > 0\}$ are assumed to be independent on the field.

For non-stationary process the equation of state has the form [11]:

$$\frac{1}{L}\frac{\partial\eta}{\partial t} = -\frac{\delta F}{\delta\eta}, \tag{2}$$

where L is the kinetic coefficient.

Spontaneous polarization $\vec{P}$ [2–5, 7, 10] or electric displacement $\vec{D}$ [12, 13] can be chosen as the order parameter η. The problem of choice of the order parameter is in detail discussed in Ref. [14]. According to variational calculus, the corner-Weierstrass-Erdmann conditions should be performed in the internal interfaces [15]. For a functional (1), these conditions reduce to the continuity of the normal component of η and its derivative with respect to the normal. If we choose the polarization as η, then, taking into account the continuity of $\vec{D}$ normal component, we obtain the continuity of $\vec{E}$ normal components. This fact contradicts to the classical Maxwell equations. Therefore, $\eta = \vec{D}$ and, as a result, we obtain an equation:

$$g\Delta\vec{D} - \frac{1}{L}\frac{\partial\vec{D}}{\partial t} = a\vec{D} + b\vec{D}^3 - \vec{E}. \tag{3}$$

It should be noted that other variants of the Eq. (3) are possible. For example, the time derivative and the power series expansion of the right-hand side of Eq. (3) may contain $\vec{P}$.

Modeling of Thin "Dead Layers"

In a cylindrical coordinate system (r, θ, z), we consider a three-layered structure shown in Fig. 1. To simplify the subsequent calculations, we exclude the layer from the consideration. To do this, we derive a formula relating the values of the electric potential φ and field $\vec{E} = -\nabla\varphi$ on the boundaries of the ferroelectric and dielectric with "dead layer". Poisson equation for the layer has the form:

$$\mathrm{div}\vec{D} = \rho, \tag{4}$$

where $\rho = \rho(r, \theta, z)$ is volume charge density The explicit form of ρ coordinate dependence is not significant in our consideration. Let us integrate both sides of Eq. (4) over z within

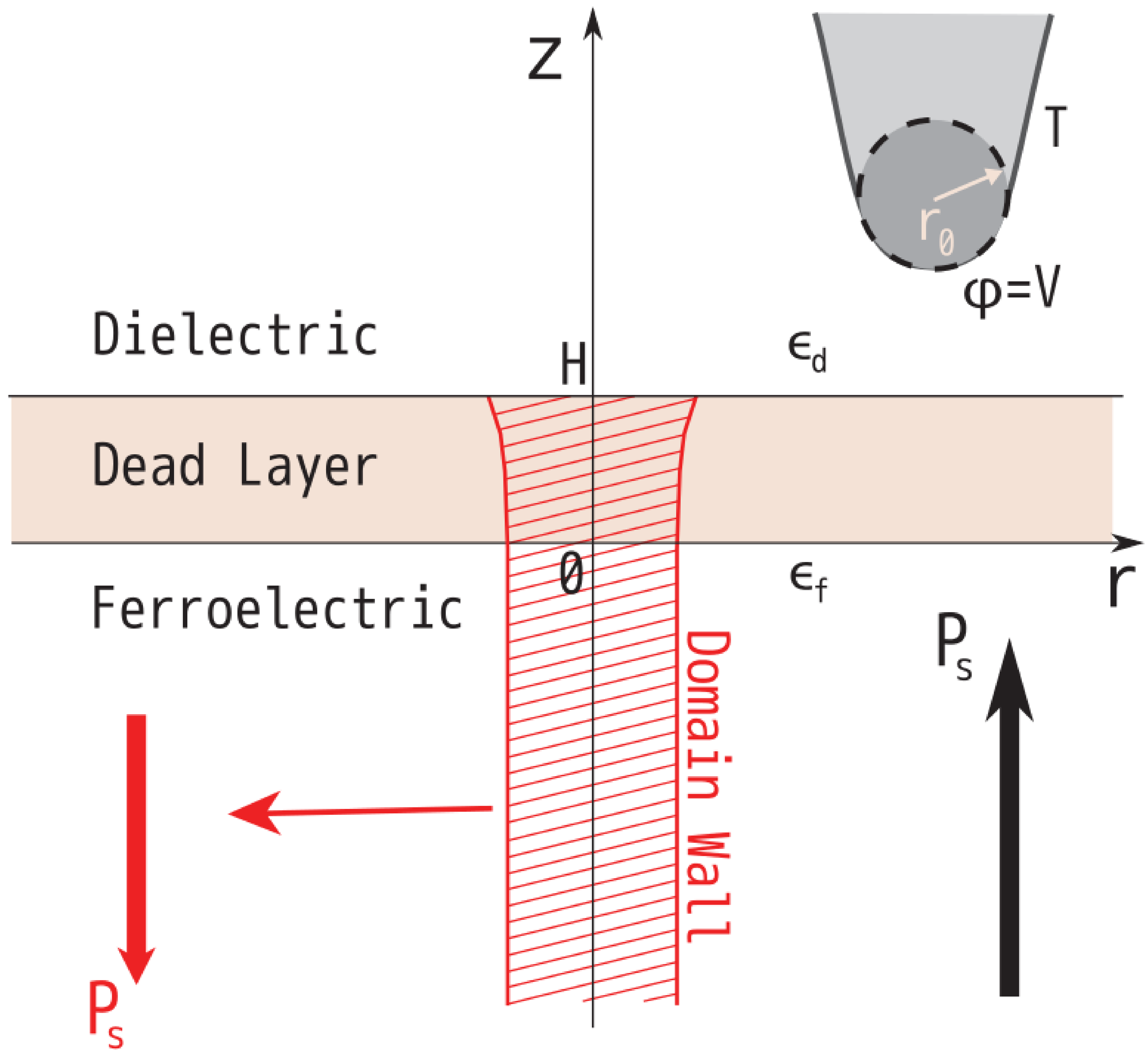

Figure 1. The schematic representation of the system under consideration—SPM tip/dead layer/ferroelectric film. The arrows inside the structure indicate the spontaneous polarization directions (Color figure available online).

the "dead layer" (i.e., from 0 to H). We get the relations for the components of $\vec{D} = \{D_r; D_\theta; D_z\}$:

$$D_z(r, \theta, H) - D_z(r, \theta, 0) = \sigma(r, \theta). \tag{5}$$

Here, the value of

$$\sigma(r, \theta) = \int_0^H (\rho(r, \theta, z) - D_1(r, \theta, z))\, dz \tag{6}$$

is the effective surface charge density and

$$D_1(r, \theta, z) = \frac{1}{r}\frac{\partial}{\partial r} r D_r + \frac{1}{r}\frac{\partial D_\theta}{\partial \theta}. \tag{7}$$

Thus, instead of the two boundary conditions for the continuity of the electric displacement we have only one condition (5). If both sides of Eq. (4) are pre-multiplied by z and then are integrated within the "dead layer", we obtain a relation between the values of the potential on the two sides of the layer:

$$\phi(r, \theta, H) - \phi(r, \theta, 0) = -H E_z(r, \theta, H) + \tau(r, \theta), \tag{8}$$

$$\tau(r, \theta) = \frac{1}{\varepsilon_0} \int_0^H z(\rho(r, \theta, z) - D_1(r, \theta, z) - P_z)\, dz. \tag{9}$$

where, ε_0 is the universal dielectric constant. Note that the integrand in Eq. (9) has a small factor z and, in addition, the integration is performed over layer with a small thickness. Consequently, the value of τ as the integral of the second term in Eq. (6), in the first approximation can be neglected.

Account of the High Dielectric Constant of Ferroelectrics

For the description simplicity, in this section we consider the linearized model of the ferroelectric (hard-ferroelectric approximation). For this, let us consider a ratio of the dielectric constants of the dielectric ε_{d} and ferroelectric ε_{f}, $s = \varepsilon_{\mathrm{d}}/\varepsilon_{\mathrm{f}}$. It is well known that for conventional ferroelectric materials this parameter varies in the range of $10^{-4} \div 10^{-2}$ [16]. The potential φ satisfies the Laplace equation $\Delta\phi = 0$ (in the half-spaces), the boundary conditions (8) and the following one:

$$\frac{\partial\varphi}{\partial z}(r, \theta, 0) = s\frac{\partial\varphi}{\partial z}(r, \theta, H) + \frac{\sigma(r, \theta) + P_{\mathrm{z}}}{\varepsilon_0\varepsilon_{\mathrm{f}}}. \tag{10}$$

According to Ref. [17], the electric field potential can be expressed by the following power series over s:

$$\varphi = \sum_{j=0}^{\infty} \varphi_j(r, \theta, z)s^j. \tag{11}$$

The substitution of Eq. (11) in Eqs. (8) and (10) and equalization of the coefficients with the same power of s leads to the system of equations. From this system, we can define $\varphi_0(r, \theta, z) = 0$ for $z<0$. From this equality, it follows that for small s the electric field distributions in the dielectric and ferroelectric can be considered independently from each other. The calculation results of the field from the exact formula and the approximate one are shown in Fig. 2. This fact significantly simplifies the calculations. To be more precise, on the first step the electric potential in the dielectric is found. Further, the electric displacement in the ferroelectric film is defined by using the known value of the electric field intensity at the interface:

$$D_{\mathrm{z}} = \varepsilon_0\varepsilon_{\mathrm{d}}E_{\mathrm{dz}} - \sigma - P_{\mathrm{z}}, \tag{12}$$

where E_{dz} is the vertical component of electric field in dielectric.

III. Mathematical Statement of the Problem and its Solution

Introduction of Dimensionless Quantities and Variables

We shall consider the one-dimensional problem, i.e. study the motion of a planar domain wall. The accuracy of this assumption is of the order of $D_{\mathrm{z}}/|\vec{D}|$. It can be shown that the inclusion of the elastic stresses in ferroelectric (arising due to the inverse piezoelectric effect and/or electrostriction) leads to a bending of the domain wall. However, at the stage of sideways growth one-dimensional model yields qualitatively correct results [10]. After the transition to a dimensionless function $u = D_{\mathrm{z}}\sqrt{-b/a}$ and dimensionless coordinates

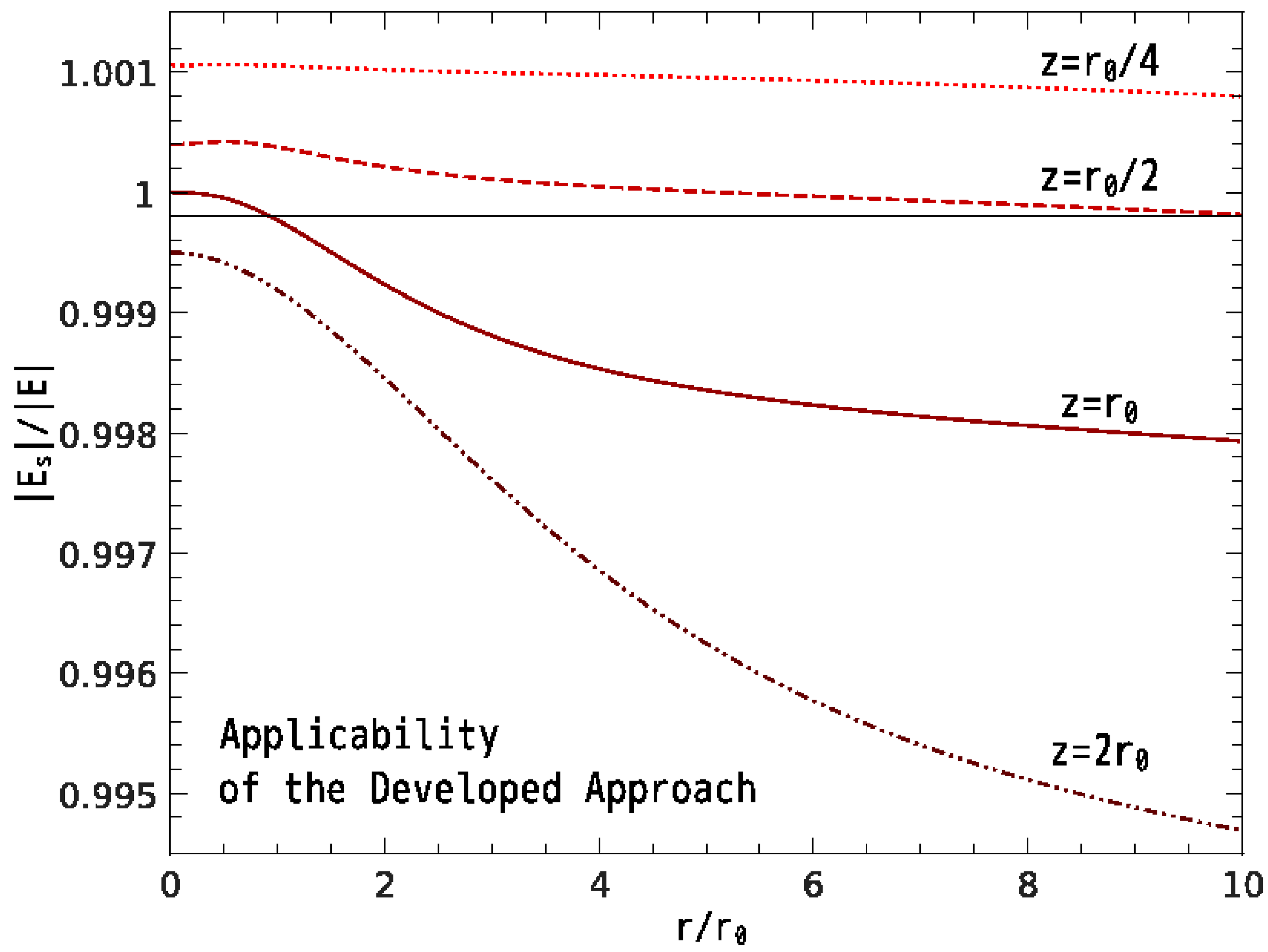

Figure 2. The absolute value of electric field as a function of the dimensionless coordinate – comparison of precise model |*E*| and suggested scheme |*E*s|. The calculation is performed for $z = \{r_0/4,\ r_0/2,\ r_0,\ 2r_0\}$. A perfect agreement between models demonstrates that electric field can be reproduced by the simplified theory with enough accuracy (Color figure available online).

$x = \sqrt{-a/g}$, $y = t/(-aL)$ we obtain the following equation:

$$\frac{\partial^2 u}{\partial x^2} - \frac{\partial u}{\partial y} = -u + u^3 - e(x, y), \tag{13}$$

where

$$e(x, y) = \frac{\sqrt{b}}{-a\varepsilon_0\sqrt{-a}}(\varepsilon_0\varepsilon_{\mathrm{d}}E_{\mathrm{dz}}(x, y) - \sigma(x, y) - P_{\mathrm{z}}(x, y)). \tag{14}$$

The main difference between Eq. (14) and considered earlier ones is an interpretation of the term $e(x,y)$. We include not only the value of the electric field normal component on the boundary, but the effective surface charge density together with the vertical component of the polarization. We shall explore Eq. (14) under the assumption of smooth change of the function $|\nabla e|/|e| \ll 1$. The initial conditions are chosen in the form: $u(x, 0) = \operatorname{sign} x$. Eq. (14) has the relaxation type and describes the transition from an unstable to a steady state, which is determined from the equation:

$$u^3 - u - e = 0. \tag{15}$$

For $e > e_{cr} = 2\sqrt{3}/9$ Eq. (15) has one real root u_3 and two complex ones: u_1, u_2 (Fig. 3). For $e < e_{cr}$ all roots of Eq. (15) $u_1 < u_2 < u_3$ are real. In the standard LGD model the value e_{cr} corresponds to the coercive field E_{coer}. Solution of Eq. (14) will also have a different form at $e > e_{cr}$ and $e < e_{cr}$.

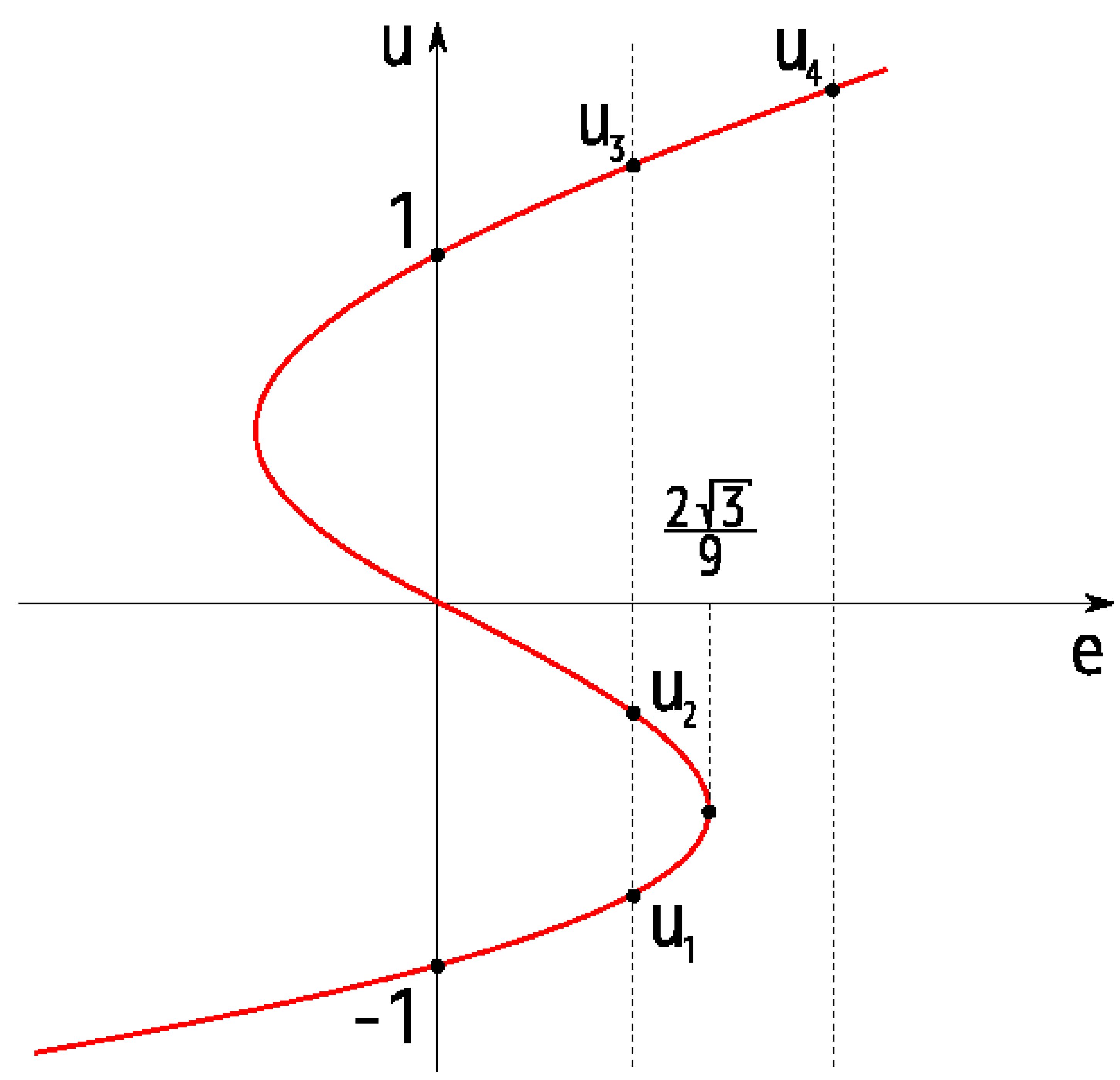

Figure 3. The roots of the cubic equation (Color figure available online).

Solution of One-Dimensional Problem

First, consider the case of $e < e_{cr}$. We seek normalized displacement as $u(x + vy)$, where v is the velocity of the domain wall. Omitting the simple mathematical calculations, we present only results based on the ideas of Ref. [18]. The velocity is the sum of two terms:

$$\begin{aligned} v &= v_0 + v_1, \\ v_0 &= \frac{(u_3 - u_1)(u_3 + u_1 - 3e)}{2\int\limits_{u_1}^{u_3} \sqrt{c + u^4/2 - u^2 - 2eu}\,du}, \quad v_1 = \frac{\int\limits_{-\infty}^{\infty} (\partial^2/\partial x^2 - v_0\partial/\partial y)\, u[X, e(x, y)]|_{X=x}\, dx}{\int\limits_{-\infty}^{\infty} (\partial u/\partial x)^2\, dx}. \end{aligned} \tag{16}$$

The function u is determined by integration of:

$$\left(\frac{\partial u}{\partial x}\right)^2 = v_0 \int\limits_{u_1}^{u} \sqrt{c + \frac{u^4}{2} - u^2 - 2eu}\,du + c + \frac{u^4}{2} - u^2 - 2eu \tag{17}$$

for $c = u_1^2/2 + 3eu_1/2$. The constant of integration c and v_0 can be found from conditions at $x \to \infty$. Component of the velocity v_0 is determined by the local properties of the field, depending on the coordinate through dependence on e. The second term in Eq. (16) is small and is proportional to the characteristic size of the inhomogeneities. However, it depends on the global properties of the field. The function $\partial u/\partial x$ is different from 0 only in a small vicinity of the domain wall. Thus, one may conclude that v_1 depends on the value of the electric field only near the domain walls. The results of calculations performed on the base of Eqs. (16) and (17) are presented in Fig. 4.

For the case of large electric field, i.e. $e > e_{cr}$, the asymptotic solution of Eq. (14) satisfying the initial conditions can be written in an implicit form:

$$y(u) = \sum_{i=1}^{3} A_i \ln\left(\frac{u_i - u}{u_i + 1}\right), \tag{18}$$

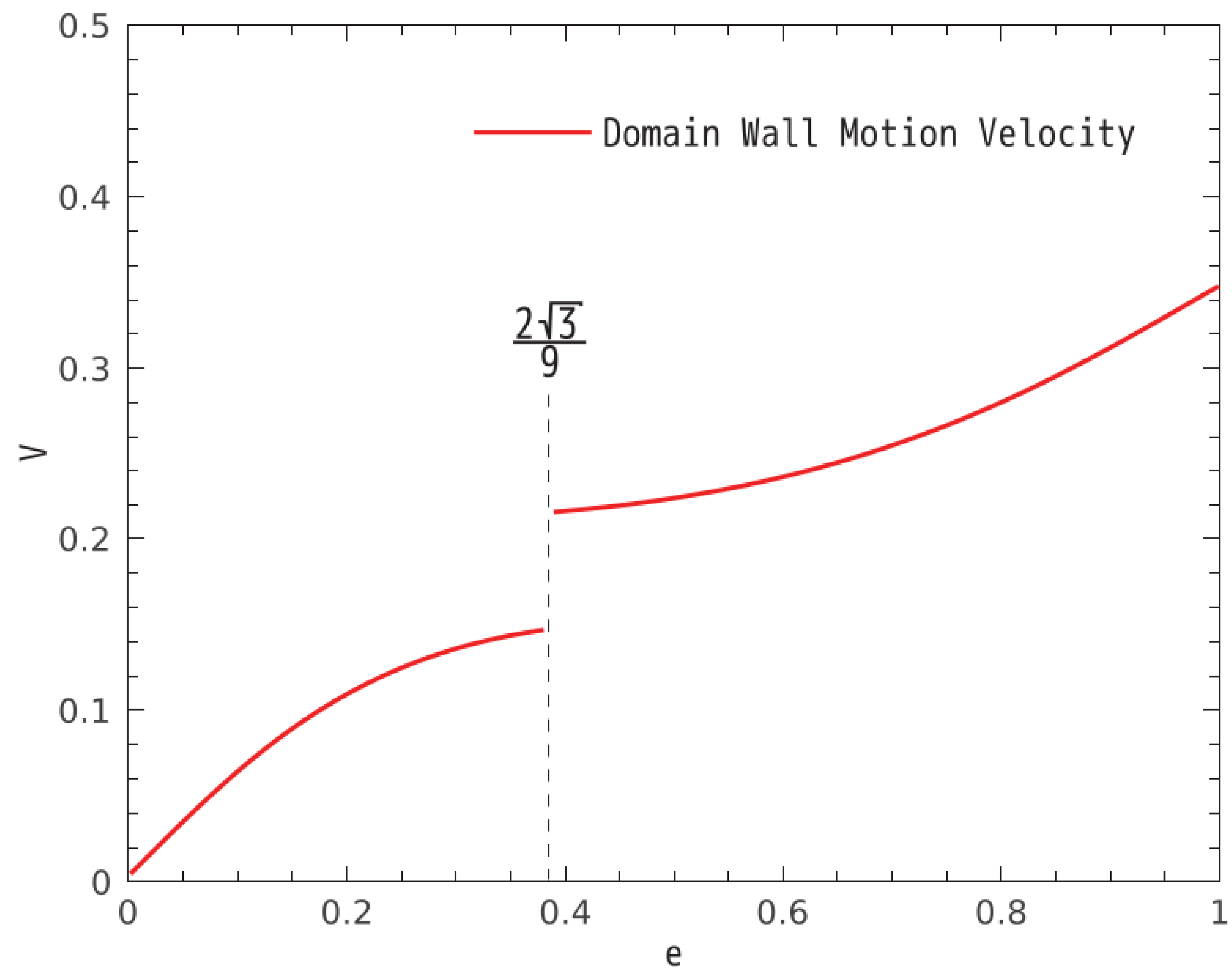

Figure 4. The domain wall motion velocity. One can clearly see that such dependence cannot be approximated by a simple function (e.g., power function) (Color figure available online).

where:

$$A_i = \frac{\partial u_i}{\partial e} = \frac{1}{3u_i^2 - 1}. \tag{19}$$

The position of the domain wall is determined from the condition $u = 0$. Differentiating the implicit function (18) on stretched time we obtain the formula for velocity:

$$v = \left(\left(\frac{\partial y(0)}{\partial e} \right)^{-1} - \frac{\partial e}{\partial y} \right) \bigg/ \frac{\partial e}{\partial x}. \tag{20}$$

Note that in this case (called spinodal decomposition) the velocity of the domain wall is determined not only by the value of the electric field e, but also its derivatives.

Influence of Effective Surface Charge Density

In our model, the effective surface charge density σ is considered as a known quantity. But, for a more precise description of the domain wall motion, Eq. (14) should be supplemented by the expression for σ. As this equation, we choose the natural generalization of the σ definition to the nonstationary case:

$$\frac{1}{L_\sigma} \frac{\mathrm{d}\sigma}{\mathrm{d}t} = \sigma_0 - \sigma. \tag{21}$$

where σ_0 is the steady value of the effective density of surface charges and L_σ is the kinetic coefficient characterizing the relaxation time of σ. As σ_0, it is possible to choose spontaneous polarization multiplied by a screening coefficient. Thus, the solution of Eq. (21) has the form:

$$\sigma = \sigma_0 + (\sigma_{\mathrm{in}} - \sigma_0) \exp(-L_\sigma t), \tag{22}$$

where σ_{in} is initial value of σ. Impact of the time dependence of σ on the velocity of the domain wall is determined by the ratio of the kinetic coefficients L_σ/L. In some cases it may be a full stop of the domain wall. Eq. (22) gives the following formula for pause time:

$$\tau = -\frac{1}{L_\sigma}\ln\left(\frac{\varepsilon_0\varepsilon_{\text{d}}E_{\text{dz}}}{2P_{\text{S}}}\right). \tag{23}$$

From analysis of this formula we conclude that the stop of the domain wall is possible only in the case of small fields, for which the argument of the logarithm of Eq. (23) is less than 1.

IV. Conclusion

In this work, we have analyzed the asymptotic solution of the one-dimensional time domain LGD problem. It is shown that the evolution of charge density with time is of great importance for the domain wall motion description. For $E > E_{\text{coer}}$, the domain motion velocity depends not only on the electric field value, but on the space and time derivatives. For small values of the electric field, the domain motion velocity can be defined as sum of two summands: the first is described via local parameters and the second is an integral characteristic. One may conclude that the dependence of the domain wall motion velocity on the external field is complicated enough to be approximated by a power or exponential function. Taking into account the dependence of effective surface charge density on time, we explain the jump-like kinetics of the domain wall observed in experiments and estimate the time of this interruption.

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References

1. R. C. Miller and G. Weinreich, Mechanism for the sidewise motion of 180° domain walls in barium titanate. *Phys. Rev. B* **117**, 1460 (1960).
2. A. N. Morozovska, S. V. Kalinin, E. A. Eliseev, V. Gopalan, and S. V. Svechnikov, Interaction of a 180° ferroelectric domain wall with a biased scanning probe microscopy tip: Effective wall geometry and thermodynamics in Ginzburg-Landau-Devonshire theory. *Phys. Rev. B* **78**, 125407 (2008).
3. E. A. Eliseev, A. N. Morozovska, G. S. Svechnikov, E. L. Rumyantsev, E. I. Shishkin, V. Y. Shur, and S. V. Kalinin, Screening and retardation effects on 180°-domain wall motion in ferroelectrics: Wall velocity and nonlinear dynamics due to polarization-screening charge interactions. *Phys. Rev. B* **78**, 245409 (2008).
4. A. N. Morozovska, G. S. Svechnikov, E. I. Shishkin, and V. Y. Shur, Domain structure formation by using scanning probe microscopy: equilibrium polarization distribution and effective piezo-electric response calculations. *Semicond. Phys. Quantum Electron. Optoelectron.* **12**, 116–124 (2008).
5. V. R. Aravind, A. N. Morozovska, S. Bhattacharyya, D. Lee, S. Jesse, I. Grinberg, Y. L. Li, S. Choudhury, P. Wu, K. Seal, A. M. Rappe, S. V. Svechnikov, E. A. Eliseev, S. R. Phillpot, L. Q.Chen, V. Gopalan, and S. V. Kalinin, Correlated polarization switching in the proximity of a 180° domain wall. *Phys. Rev. B* **82**, 024111 (2010).

6. J. Wang, Y. Xia, L.-Q. Chen, and S.-Q. Shi, Effect of strain and deadlayer on the polarization switching of ferroelectric thin film. *J. Appl. Phys.* **110**, 114111 (2011).
7. A. N. Morozovska, E. A. Eliseev, S. L. Bravina, and S. V. Kalinin, Landau-Ginzburg-Devonshire theory for electromechanical hysteresis loop formation in piezoresponse force microscopy of thin films. *J. Appl. Phys.* **110**, 052011 (2011).
8. V. M. Fridkin, Domains and switching in ferroelectricity. *Ferroelectrics* **426**, 139–141 (2012).
9. V. Fridkin, M. Kuehn, and H. Kliem, The Weiss model and the Landau–Khalatnikov model for the switching of ferroelectrics. *Physica B* **407**, 2211–2214 (2012).
10. A. K. Tagantsev, L. E. Cross, and J. Fousek, *Domains in ferroic crystals and thin film*. (Springer, New York 2010).
11. L. D. Landau and I. M. Khalatnikov, About anomalous sound attenuation near the phase transition of second order. *Dokl. Akad. Nauk SSSR* **96**, 465–468 (1954).
12. A. N. Morozovska and E. A. Eliseev, Modelling of dielectric hysteresis loops in ferroelectric semiconductors with charged defects. *J. Phys. Condens. Matter* **16**, 8937 (2004).
13. A. N. Morozovska and E. A. Eliseev, Theoretical description of ferroelectric and pyroelectric hystereses in the disordered ferroelectric-semiconductor films. *J. Appl. Phys.* **100**, 014109 (2006).
14. A. K. Tagantsev, Landau Expansion for ferroelectrics: Which variable to use?, *Ferroelectrics* **375**, 19–27 (2008).
15. O. Bolsa, *Lectures on the calculus of variations*. (Dover, New York 1961).
16. A. Tagantsev, V. Sherman, K. Astafiev, J. Venkatesh, and N. Setter, Ferroelectric materials for microwave tunable applications. *J. Electroceram.* **11**, 5–66 (2003).
17. F. Olver, *Asymptotics and Special Functions*. (Academic Press, New York 1974).
18. J. C. Luke, A perturbation method for nonlinear dispersive wave problems. *Proc. R. Soc. London, Ser. A* **252**, 403–412 (1966).