Ferroelectric charged domain walls in an applied electric field

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The interaction of electric field with charged domain walls in ferroelectrics is theoretically addressed. A general expression for the force acting per unit area of a charged domain wall carrying free charge is derived. It is shown that, in proper ferroelectrics, the free charge carried by the wall is dependent on the size of the adjacent domains. As a result, the mobility of such domain wall (with respect to the applied field) is sensitive to the parameters of the domain pattern containing this wall. The problem of the force acting on a charged planar 180° domain wall normal to the polarization direction in a periodic domain pattern in a proper ferroelectric is analytically solved in terms of Landau theory. In small applied fields (in the linear regime), the force acting on the wall in such pattern increases with decreasing the wall spacing. It is shown that the domain pattern considered is unstable in a defect-free ferroelectric. The poling of a crystal containing such pattern, stabilized by the pinning pressure, is also considered. Except for a special situation, the presence of charge domain walls makes poling more difficult. The results obtained are also applicable to zigzag walls under the condition that the zigzag amplitude is much smaller than the sizes of the neighboring domains.

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I. INTRODUCTION

Typically, at domain walls in ferroelectrics, the normal component of electrical polarization is conserved with high accuracy. This is the so-called condition of electrical compatibility.[1] When a ferroelectric behaves as a perfect insulator, any appreciable violation of this condition will imply the appearance of bound charge at the wall and a macroscopic electric field in the adjacent domains. Such a field, at best, can strongly increase the energy of the system, but very often this field is expected to be strong enough to fully suppress the ferroelectricity in the sample. At the same time, the walls with an essential violation of the electrical compatibility, the so-called charged domain walls, have been observed in a number of materials such as lead titanate $PbTiO_3$ (PT),[2–4] lead zirconate titanate $Pb[Zr_xTi_{1-x}]O_3$ (PZT),[5] lead germanate $Pb_5Ge_3O_{11}$ (PGO),[6,7] and lithium niobate $LiNbO_3$.[8,9] It is believed that such walls can exist due to the compensation of their bound charge by ions[8,9] or free electrons.[2–4] Here, the situation with the electron compensation is of special interest. Specifically, as it was recognized many years ago, the concentration of free carriers in properly compensated domain walls can readily achieve the metallic level.[10,11] Thus, one can treat such a wall as a highly conductive interface. In principle, one can tune its conductivity by controlling the wall orientation[12] and change its position in the crystal by the application of a dc electric field. In this context, mobility of charged domain walls is an important issue. Additionally, this issue deserves attention in view of poling samples containing charged domain walls.

The interaction of electric field with a charged domain wall has not been completely understood and theoretical results are limited. Here, one can mention only a classical paper by Landauer[13] and a recent paper by Mokrý *et al.*[14] Landauer indicated that the compensation of the bound charge on a wall will lead to a reduction of the pressure acting on it in the presence of a dc electric field, whereas Mokrý *et al.* quantitatively described this effect in terms of Landau theory. A closer analysis of the problem, however, shows that it misses a more involved theoretical treatment. For example, a straightforward application of the results by Mokrý *et al.*[14] to the case of a zigzag charged wall leads to a paradox. Also a free charge density on a domain wall was considered in Ref. 14 as an independent parameter, while in reality this charge is a function of the sizes of the neighboring domains.

The goal of this paper is to revisit the problem of forces acting at charged domain walls in an electric field. The paper is organized as follows. In Sec. II, we discuss and solve the aforementioned paradox. Here, we obtain the general expression for the local force acting on the domain wall. It is shown that not only the normal component of the force (treated in Ref. 14), but also the tangential one should be considered. In Sec. III, we consider the force acting on a charged planar 180° domain wall normal to the polarization direction in a periodic domain pattern in a proper ferroelectric. We have shown that, for a fixed applied field, the force acting on the domain wall is a function of the structure period. The results obtained are applied to the analysis of poling of such a structure. In Appendixes, supporting calculations for Sec. II are presented.

II. LOCAL FORCE DENSITY ON A FERROELECTRIC DOMAIN WALL

In this section, we will revisit the problem of the force acting on a charged domain wall due to an electric field. After Mokrý *et al.*,[14] who originally addressed this problem, we will use the following model. First, the internal structure of domain wall is neglected. Second, the free charge is moving together with the domain wall. Third, we will not consider the forces arising due to an imbalance of the elastic energy, which is a suitable approximation for nonferroelastic domain walls.

Mokrý *et al.*[14] obtained a relation linking the pressure acting on the wall (the surface density of the normal component of the force) with values of polarization $\vec{P}$, electric field $\vec{E}$ in the

adjacent domains, and surface density σ_f of the free charge carried by the wall as

$$p = [[\Phi(P)]] - \widehat{E}_i\,([[P_i]] - \sigma_f n_i)\,. \tag{1}$$

Here, Φ is a thermodynamic function characterized by the differential $d\Phi = E_i dP_i$, $[[Z]] = Z^{(2)} - Z^{(1)}$ denotes the jump of the quantity Z on the domain wall, $\widehat{E} = (E^{(1)} + E^{(2)})/2$, the upper suffixes are designating the domains, and n_k is a unit vector, normal to the domain wall, directed from domain (1) to domain (2). The Einstein summation convention is used. The positive sign of p corresponds to the force acting on the domain wall from domain (1) to domain (2).[15]

However, a straightforward application of this result for the case of the force acting on charged zigzag walls (Fig. 1), which are typically observed in experiments,[2–4,7] leads to a paradox. This can be demonstrated in a simple model of a fully compensated domain wall in the hard ferroelectric approximation used by Mokrý *et al.*[14] In this approximation, the electrical displacement $\vec{D}$ inside the domains is presented as a sum of the constant spontaneous polarization $\vec{P}_0$ and a linear dielectric response to the electric field with permittivity $(\varepsilon_f)_{ij}$:

$$D_i = (\varepsilon_f)_{ij} E_j + P_{0i}. \tag{2}$$

We will consider the zigzag wall in a parallel plate ferroelectric capacitor with the polarizations inside the domains perpendicular to the electrodes (Fig. 1). The bottom electrode of the capacitor is grounded and a constant potential $-U$ is applied to the top electrode.

First, let us obtain the force acting on this wall using Eq. (1). The free charge density on a fully compensated head-to-head domain wall, which is equal to the jump of the normal component of the spontaneous polarization, reads as

$$\sigma_f = [[P_i]]\, n_i. \tag{3}$$

By substituting Eq. (3) into Eq. (1), one can get the pressure acting on an inclined flat segment of the zigzag domain wall as

$$p = [[\Phi(P)]] - \widehat{E}_i\left([[P_i]] - \left[\left[P_j\right]\right] n_j n_i\right). \tag{4}$$

In the considered model $[[\Phi(P)]] = 0$, $\widehat{E}_i = E_0 l_i$, $[[P_i]] = -2P_0 l_i$, where $l_i = E_i/E_0$ is a unit vector in the direction of the electric field. Thus, Eq. (4) can be rewritten as

$$p = 2P_0 E_0 (1 - l_i n_i l_j n_j) > 0. \tag{5}$$

According to Eq. (5), the z component of the force acting on each segment of the zigzag domain wall is directed from domain (1) to domain (2), thus the total force acting on the wall is nonzero.

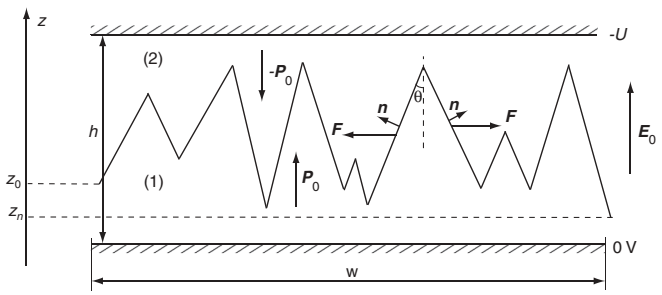

FIG. 1. Schematic of zigzag domain wall in ferroelectric capacitor. The polarization vectors inside domains are perpendicular to the electrodes. The bottom electrode is grounded and a constant potential $-U$ is applied to the top electrode. Directions of the electric field and polarizations inside the domains are shown with arrows. The z axis is parallel to the electric field.

On the other hand, we can obtain this force directly from the principle of virtual displacement. According to this principle, the work δW done by the external force F is equal to the variation of the proper thermodynamic function δG during a virtual displacement δu:

$$\delta W = F\delta u = \delta G. \tag{6}$$

For considered system, in the hard ferroelectric approximation, the thermodynamic function can be defined as[14]

$$G = \int_V \frac{1}{2}(\varepsilon_f)_{ij} E_i E_j dV + U Q, \tag{7}$$

where Q is the charge on the top electrode, and E is the electric field. Due to the full compensation of the bound charge on the zigzag wall, the electric field inside the ferroelectric is equal to U/h and does not change with the virtual displacement. This means that the variation of the thermodynamic function δG, produced by the virtual displacement δu, is given only by the variation of the charge on the top electrode δQ. Since the zigzag wall is electroneutral, the virtual displacement of this wall will not affect the charges on the electrodes (see Fig. 1), thus the thermodynamic function G will not change with the virtual displacement. This implies that the force acting on the wall is zero. Thus, we are in conflict with the result obtained from Eq. (1).

The above paradox can be resolved if we take into account the fact that the local force is not necessarily perpendicular to the domain wall. Thus, to calculate the resultant force, acting on an element of the domain wall, it is not sufficient to know the pressure acting on it. The tangential component of the force (with respect to the domain wall) should be taken into account. As we will see later, in the case of the fully compensated zigzag wall, the consideration of all components of the force will give the resultant force equal to zero.

Let us obtain the general expression for the force density on the domain wall using the principle of virtual displacement. We can not use the formula for the generalized stress tensor from textbook by Landau and Lifshitz,[16] as was done in Ref. 14. In fact, no derivation of this formula is available. In addition, one can show that it leads to results which are in contradiction with those obtained with the principle of virtual displacement (see Appendix A).

In order to obtain a general formula for the local force density on domain wall, we follow the principle of virtual displacements, as applied to a domain wall of an arbitrary shape. We will consider the electric field created by conductors at fixed potentials. We will use the thermodynamic function G of the general form[14]

$$G = \int_V \left[\Phi(P) + \frac{1}{2}\varepsilon_b E^2\right] dV - \sum_i \varphi_E^{(i)}\, Q_E^{(i)}, \tag{8}$$

where the first term represents the part of the thermodynamic function $\Phi(P)$, associated with the ferroelectric part of

polarization P, and the electric field energy integrated over the volume V of the ferroelectric; the second term represents the subtracted work of the electric sources ($\varphi_E^{(i)}$ and $Q_E^{(i)}$ are the potential and charge on the i-th conductor). Here, ε_b is the background permittivity, which was for simplicity taken as isotropic. Thus, in our notation, the vector of electric displacement reads as

$$D_i = \varepsilon_b E_i + P_i. \tag{9}$$

A straightforward analysis in terms of virtual displacement, given in Appendix B, leads to the following expression for the force acting on a unit area of the domain wall:

$$f_k = [[\Phi(P)]]\, n_k - \widehat{E}_i\, [[P_i]]\, n_k + \widehat{E}_k \sigma_f. \tag{10}$$

The first two terms in Eq. (10) are the same as for the neutral domain wall. The third term is the Coulomb force acting on the free charge on the domain wall. We can rewrite Eq. (10) as the sum of the tangential and normal components of the force with respect to the domain wall as

$$f_k = \{[[\Phi(P)]] - \widehat{E}_i([[P_i]] - \sigma_f n_i)\} n_k + \{\widehat{E}_k - \widehat{E}_j n_j\, n_k\}\sigma_f. \tag{11}$$

The first term in the curly brackets is the pressure acting on the domain wall, which was obtained earlier by Mokrý *et al.*,[14,17] given by Eq. (1).

The second term in Eq. (11) represents the tangential component of the force with respect to the domain wall

$$f_{||} = \widehat{E}_{||}\sigma_f, \tag{12}$$

where $\widehat{E}_{||k} = \widehat{E}_k - \widehat{E}_j n_j\, n_k$ is the tangential component of the average electric field with respect to the domain wall. The force $f_{||}$ is the tangential component of the Coulomb force acting on the free charges on the wall.

Let us use the above result to find the force acting on a fully compensated zigzag domain wall, the situation where we have come across the paradox. As before, we consider the problem in the hard ferroelectric approximation. To exclude an edge effect, we consider a parallel plate capacitor with the thickness h, which is much smaller than the capacitor width w:

$$h/w \ll 1. \tag{13}$$

In this model, we will deal with the force per unit area of the electrode in the limit of infinitely wide capacitor instead of the total force acting on the wall.

Using Eqs. (3) and (10), one finds the force acting on a flat segment of the domain wall in the form

$$F_k = f_k S = (-\widehat{E}_i\, [[P_i]]\, n_k + \widehat{E}_k\, [[P_i]]\, n_i) S, \tag{14}$$

where S is the area of the wall segment.

In the geometry of the considered problem (see Fig. 1), Eq. (14) can be simplified as

$$F_k = f_k S = 2P_0E_0[n_k - l_k(l_i n_i)]S. \tag{15}$$

The vector $n_k - l_k(l_i n_i)$ is the component of the n_k parallel to the electrodes. Thus, the force F_k is parallel to the capacitor electrodes, and in accordance with the definition of n_k acting from domain 1 to domain 2. The absolute value of this force is proportional to the area of the segment projected on the z axis, i.e., $|F_k| = 2P_0E_0L|z_i - z_{i-1}|$, where the z axis is directed perpendicular to the electrodes, z_i are the coordinates of the zigzag vertexes, and L is the capacitor width in the direction, perpendicular to the figure plane (Fig. 1). The direction of this force depends on the sign of the angle θ, i.e., on the sign of the difference $z_i - z_{i-1}$. Thus, the absolute value of the total force acting on the zigzag wall containing n segments can be found as the sum the the forces acting on each segment

$$|F| = 2P_0E_0L\left|\sum_{i=0}^{n}(z_i - z_{i-1})\right| = 2P_0E_0L|z_n - z_0|. \tag{16}$$

The force obtained depends only on the coordinates of the vertexes at the edges of the capacitor. Considering the density of this force per unit area of the electrode, we get

$$\left|\frac{F}{S_{el}}\right| = \frac{2P_0E_0L\,|z_n - z_0|}{Lw} \leqslant \frac{2P_0E_0h}{w}. \tag{17}$$

In the considered model, the fraction h/w and, therefore, the force density $|F/S_{el}|$ vanishes. Thus, the total force acting on the wall is zero, and the above paradox is resolved. The fact that this force is zero is not just a trivial consequence of the electrical neutrality of the domain wall because an interaction between electric field and a domain wall includes not only simple electrostatic forces acting on the charge on a domain wall, but also the ponderomotive forces. For example, the force acting on the 180° neutral domain wall is equal to $2P_0E_0$, while there is no electric charge on this wall.

Concluding this section, we would like to discuss the applicability of Eq. (10) to the case of a partially compensated zigzag wall. To calculate the force acting on such a wall, one should know the electric field and the polarization on the both sides of the wall and apply Eq. (10) locally. In general, this is a tough mathematical problem. However, the result can be readily obtained in the case where the zigzag amplitude d is much smaller than the domain size h (see Fig. 2). We can consider such a zigzag wall together with a layer of thickness

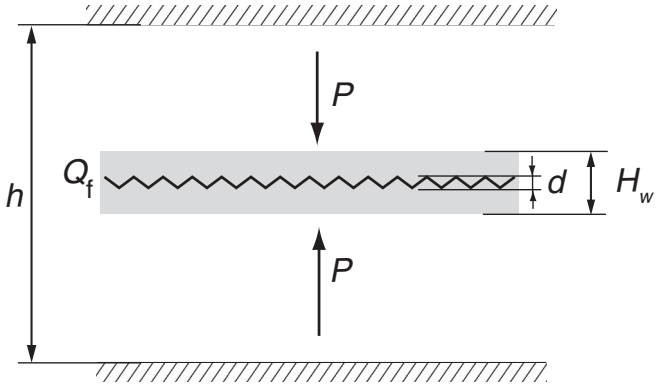

FIG. 2. Schematic of zigzag domain walls in a ferroelectric capacitor. The polarizations inside domains are perpendicular to electrodes. The zigzag amplitude d is much smaller than the domain size. This wall can be considered together with a layer around it of thickness H_w ($d \ll H_w \ll h$), which is shown with the dark region on the figure. The polarizations inside the bulk of the domains, the free electric charges on the walls, and the force acting on the walls are the same for the zigzag wall and the equivalent flat wall.

H_w, $d \ll H_w \ll h$, containing it. This layer is equivalent to a flat wall: they have the same jump of polarization and the same free charge, which create the same electric field in the bulk of the domains. The virtual displacement of the walls will lead to the same changes of the charges on the electrodes and the same changes in the fractions of the domains. Thus, the total forces acting on the zigzag wall and the equivalent flat wall should be the same in the limit of $d/h \ll 1$. This enables the use of Eq. (10) for the calculation the force acting on the walls of this type.

III. FORCE ON A DOMAIN WALL IN A PERIODIC PATTERN

In the previous section, we obtained the force acting on the domain wall, carrying an arbitrary free charge density σ_f. The calculation of such force requires the knowledge of this charge. An essential interesting feature of this problem is that σ_f is dependent, in a unique way, on other parameters of the system. This makes it possible to calculate the force, acting on a charged wall in a domain pattern due to the application of an external electric field. We will do this for the case of a periodic domain pattern with charged walls (see Fig. 3). We will be interested in the situation, typical for ferroelectrics,[18] where the screening of the bound charge in the wall is in the nonlinear regime, i.e., the carrier concentration inside the domain wall is much higher than the homogeneous carrier concentration in the material. We will consider a pattern with domains of the same thicknesses $H/2$, where H is the structure period, having planar walls perpendicular to the spontaneous polarization.

As was shown in Refs. 18 and 19, the charged wall in a finite sample is not completely compensated. The net charge on the wall is proportional (due to the Gauss law) to the electric field inside the adjacent domains. In a pattern containing charged domain walls, this field is not zero, even in the absence of external bias field. For the situation considered,

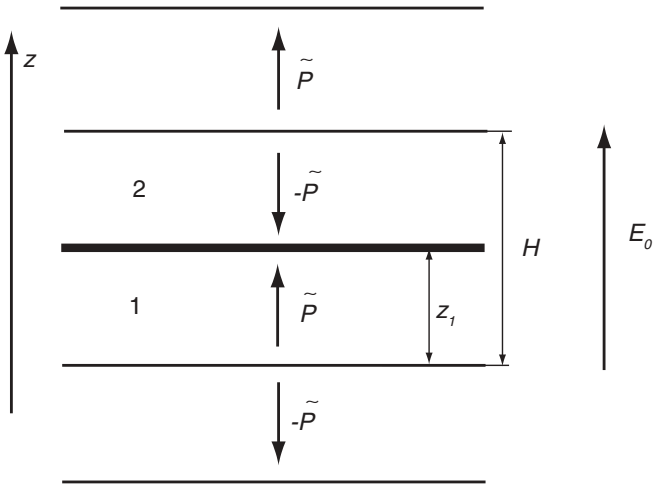

FIG. 3. Schematic of the periodic structure with head-to-head and tail-to-tail 180° domain walls with period H. Domain walls are perpendicular to the polarization inside the domains $\tilde{P}$, which is different from the spontaneous polarization due to the depolarizing field. The wall between the domains 1 and 2 considered in calculations is marked by a bold line.

where the screening regime in the wall is nonlinear, this field can readily be evaluated. Indeed, in this regime, the free carrier density, needed for the polarization screening, can be provided exclusively by a band bending. Obviously, such band bending implies a voltage difference between the neighboring head-to-head and tail-to-tail domain walls, which is about E_g/q, where E_g is the band gap of the ferroelectric and q is the absolute value of the electron charge.[11,18] Finally, the absolute value of the corresponding electric field inside a domain of the thickness $H/2$ is

$$\tilde{E} = \frac{2E_g}{qH}. \tag{18}$$

Actually, it can also be viewed as a partially compensated depolarizing field. Due to this field, the polarization inside the domains is reduced, compared to the nominal spontaneous polarization of the ferroelectric. For a ferroelectric with a second-order phase transition, where the polarization response is given by the equation of state $E = \alpha P + \beta P^3$, the reduced polarization $\tilde{P}$ can be found as the negative solution to the following equation:

$$\alpha\tilde{P} + \beta\tilde{P}^3 = \frac{2E_g}{qH}. \tag{19}$$

A. Pressure on the wall between domains of equal sizes

In the considered configuration, the tangential component of the force, given by Eq. (12), is zero, whereas the pressure, given by Eq. (1), is nonzero. Considering the domain wall between domains 1 and 2, one can rewrite Eq. (1) as follows:

$$p = \Phi_2 - \Phi_1 - \tfrac{1}{2}(E_1 + E_2)(P_2 - P_1 - \sigma_f). \tag{20}$$

Here, the electric field and polarization are considered positive if the corresponding vectors are directed along the z axis (Fig. 3), i.e., from domain 1 to domain 2. It follows from the form of Eq. (20) that the absolute values of the pressure acting on the head-to-head and tail-to-tail walls are equal, whereas the corresponding forces are acting in opposite directions. Thus, it suffices to calculate the pressure on a head-to-head wall. Such wall is shown in Fig. 3 by a bold line.

The general formula for the pressure linear in a small applied electric field E_0 can be obtained. Using Eq. (20) and expressing $E_0 = U/h$ as

$$E_0 = (E_1 + E_2)/2, \tag{21}$$

one finds

$$\begin{aligned} p &= \left(\frac{\partial\Phi_2}{\partial E} - \frac{\partial\Phi_1}{\partial E}\right) E_0 - \frac{1}{2}(E_1 + E_2)(P_2 - P_1 - \sigma_f) \\ &= [(E_2 - E_1)\,\chi - (P_2 - P_1 - \sigma_f)]\,E_0, \end{aligned} \tag{22}$$

where $\chi = \partial P/\partial E|_{P=\tilde{P}}$ is a susceptibility of a ferroelectric at $P = \tilde{P}$.

The Poisson equation for considered configuration has the form [see Eq. (9) for the definition of electric displacement]

$$\sigma_f = \varepsilon_b E_2 + P_2 - \varepsilon_b E_1 - P_1. \tag{23}$$

By combining Eqs. (22) and (23), we finally get the pressure as

$$p = 2\varepsilon_f \tilde{E} E_0, \tag{24}$$

where

$$\varepsilon_f = \varepsilon_b + \chi \tag{25}$$

is the permittivity of the ferroelectric at $P = \tilde{P}$. This is a general formula which is suitable disregarding the order of phase transition in the ferroelectric.

The absolute value of the electric field inside the domain $\tilde{E}$ in turn can be found from Eq. (18). Thus, from Eq. (24), we obtain

$$p = \frac{4\varepsilon_f E_g}{qH} E_0. \tag{26}$$

One should note that in contrast to the case of neutral domain wall, the main part of this pressure is coming from the term $\Phi_2 - \Phi_1$ in Eq. (20), but not from the electrostatic term $-\frac{1}{2}(E_1 + E_2)(P_2 - P_1 - \sigma_f)$.

It is instructive to compare the pressure, given by Eq. (24), for the case where the polarization inside the domains is close to the bulk spontaneous polarization P_0 with the pressure on the neutral domain wall $2P_0E_0$. For simplicity, we consider the case of a second-order phase transition. Physically, the condition that the polarization inside domains is close to P_0 means that the depolarizing field inside domains $\tilde{E}$ is much smaller than the thermodynamic coercive field $E_{coer} = [2/(3\sqrt{3})]|\alpha|P_0$. In this case, neglecting the difference between $\tilde{P}$ and P_0 and using Eq. (24) and the relation $\varepsilon_f \approx 1/(2|\alpha|)$, one finds $p/(2P_0E_0) \approx \varepsilon_f \tilde{E}/P_0 \approx 0.2\tilde{E}/E_{coer}$. Since, in the considered situation, the depolarizing field inside the domain $\tilde{E}$ is much smaller than E_{coer}, the pressure on the charged wall is expected to be much smaller than the pressure on the corresponding neutral wall.

The situation where the depolarizing field approaches the thermodynamic coercive field corresponds to a polarization instability inside domains manifesting itself in a divergency of the permittivity ε_f. This formally implies [via Eq. (24)] an unlimited increase of the pressure acting on the wall when the domain period H approaches a certain critical value. This academically interesting situation is, however, difficult to attain experimentally.[20] For this reason, in this paper we will further discuss only the situation where the ferroelectric inside domains is far from the aforementioned instability.

To calculate the terms in the pressure nonlinear in E_0, we need to know equations of state, which links polarizations and electric fields inside the domains. We will find the pressure on a domain wall for ferroelectric with a second-order phase transition. In this case, electric fields inside the domains can be calculated using the following equations of state:

$$E_1 = \alpha P_1 + \beta P_1^3, \tag{27}$$

$$E_2 = \alpha P_2 + \beta P_2^3. \tag{28}$$

Using Eqs. (21), (23), (27), and (28), we can present the polarizations inside the domains as a Taylor expansion with respect to E_0 as follows:

$$P_1 = \tilde{P} + \chi E_0 + \eta E_0^2, \tag{29}$$

$$P_2 = -\tilde{P} + \chi E_0 - \eta E_0^2, \tag{30}$$

where

$$\chi = \frac{1}{\alpha + 3\beta\tilde{P}^2} \tag{31}$$

is the susceptibility of the ferroelectric at $P = \tilde{P}$ and

$$\eta = \frac{-3\beta\tilde{P}}{(\alpha + 3\beta\tilde{P}^2)^2(\alpha + 3\beta\tilde{P}^2 + 1/\varepsilon_b)} = \frac{-3\beta\tilde{P}\chi^2}{1/\chi + 1/\varepsilon_b}. \tag{32}$$

By substituting the polarization from Eqs. (29) and (30) into Eq. (20) [where for a ferroelectric with a second-order phase transition $\Phi = (\alpha/2)P^2 + (\beta/4)P^4$] and keeping terms up to the third power with respect to E_0, one finds

$$p = \frac{\varepsilon_f}{\varepsilon_b}(2\tilde{P} + \sigma_f)E_0 - 2\beta\tilde{P}\chi^3 E_0^3. \tag{33}$$

Using Eq. (33) and Poisson equation (23), we can find the total pressure, including nonlinear term, as

$$p = \frac{4\varepsilon_f E_g}{qH} E_0 - 2\beta\tilde{P}\chi^3 E_0^3. \tag{34}$$

Here, χ and ε_f should be calculated for the reduced values of the polarization inside domains. The term linear in E_0 in the right-hand side of Eq. (34) was obtained earlier in the general case [see Eq. (26)]. The term nonlinear in E_0 has the opposite sign with respect to the linear one. For the structure with large domains, where the depolarizing field is small and $\tilde{P} \to P_0$, it leads to the nonlinear pressure obtained by Mokrý *et al.*[14]:

$$p = -(\chi^2/P_0)E_0^3, \tag{35}$$

where χ is the susceptibility at $P = P_0$.

B. Polling a sample with charged domain walls

In this section, we will consider the problem of poling a sample, where the periodical structure with charged domain walls shown in Fig. 3 was formed. We will consider the case of a small external field, where we can take into account only the part of the pressure given by Eq. (26), which is linear in E_0. For the model developed in the previous section and used in the present one, the free carrier concentration in the domain wall is close to metallic. This means that, for a realistic concentration of the trapping centers, the main part of the free carriers in the wall is not trapped, having high mobility and, thus, being able to easily follow the domain wall in its motion. In a realistic situation, the time needed for the free carriers to transfer between neighboring domain walls (which depends on the conductivity of the material and domain size) is expected to be large compared to the time of poling. That means that we can consider a model where the free charge carried by a domain wall remains constant, when the domain wall moves away from its position in the pattern with domains of equal thicknesses.

Defects which always exist in real materials lead to the domain-wall pinning. The pinning acts as dry friction in mechanics. When we apply a small pressure on a domain wall, its position is stabilized because of the pinning. To start the domain-wall motion, the applied pressure, given by Eq. (26), should be equal to the maximal "stabilizing" pinning pressure

p_{pinning}:

$$p = p_{\text{pinning}}. \tag{36}$$

Equations (36) and (26) enable us to find the electric field needed to start the motion of the walls from the initial positions in the equidistant domain pattern. However, the field, which is necessary to start the domain-wall motion, may not be sufficient for the poling of ferroelectric. To be sure that Eq. (36) represents a criterion of successful poling, we should check that the pressure p is greater or equal to the pinning pressure at any position of the wall after it starts moving. If the pining pressure does not depend on the position of the domain wall, successful poling at a given external electric field occurs, when the ponderomotive pressure acting at the wall in a shifted position is larger than that in the original position. In reality, one expects the pining pressure to be larger in the original position because of the defect accumulations at the wall during the time before the poling, which is the situation often observed experimentally.[7] It is clear that for this situation the aforementioned criterion of successful poling holds as well.

Let us check if the criterion of successful poling is met by our system, schematically depicted in Fig. 3. Since the absolute value of the pressure acting on the head-to-head and tail-to-tail walls is equal, whereas the corresponding forces are acting in the opposite directions, the period of the pattern will not be affected by the application of an electric field. Thus, when discussing poling we can address just one head-to-head wall moving between two tail-to-tail walls spaced by a time-independent distance H (see Fig. 3).

Let us first consider how the polarizations P_1 and P_2 and electric fields E_1 and E_2 inside the domains will change after a domain wall is displaced from its initial position. Differentiating the Poisson equation (23), and taking into account that the free charge on the wall remains constant, we get

$$\left(\varepsilon_b \frac{\partial E_2}{\partial P_2} + 1\right) \frac{\partial P_2}{\partial z_1} = \left(\varepsilon_b \frac{\partial E_1}{\partial P_1} + 1\right) \frac{\partial P_1}{\partial z_1}. \tag{37}$$

Here, $\partial E/\partial P \approx \chi^{-1} \approx \varepsilon_f^{-1}$. Thus, $\varepsilon_b(\partial E/\partial P) \approx \varepsilon_b/\varepsilon_f \ll 1$ and we can conclude that

$$\frac{\partial P_1}{\partial z_1} \approx \frac{\partial P_2}{\partial z_1}. \tag{38}$$

The sign of the derivatives in Eq. (38) can be obtained from the relation between average field E_0 applied to the ferroelectric and fields inside the domains E_1 and E_2:

$$E_1 z_1 + E_2(H - z_1) = E_0 H. \tag{39}$$

By differentiating this equation, we obtain

$$\frac{\partial E_1}{\partial z_1} z_1 + \frac{\partial E_2}{\partial z_1}(H - z_1) = -E_1 + E_2. \tag{40}$$

In the case of the head-to-head wall considered here (see Fig. 3) $E_1 < 0$, $E_2 > 0$, implying that the expression in the right-hand side in Eq. (40) is positive. Thus, the left-hand side expression in Eq. (40) is also positive and we obtain

$$\frac{\partial E_1}{\partial z_1} z_1 + \frac{\partial E_2}{\partial z_1}(H - z_1) = \frac{\partial E_1}{\partial P_1}\frac{\partial P_1}{\partial z_1} z_1 + \frac{\partial E_2}{\partial P_2}\frac{\partial P_2}{\partial z_1}(H - z_1) > 0. \tag{41}$$

Here, $\partial E_1/\partial P_1$ and $\partial E_2/\partial P_2$ are positive, the domain widths z_1, $H - z_1$ are also positive. Thus, it follows from Eqs. (41) and (38) that

$$\partial P_1/\partial z_1 \approx \partial P_2/\partial z_1 > 0. \tag{42}$$

In the following, we will consider the positive displacement of the domain wall, i.e., the size of the domain 1, z_1, increases during this displacement. This situation corresponds to the external field applied from domain 1 to domain 2 ($E_0 > 0$). In this case, it follows from Eq. (42) that the polarization in domain 1 is positive, and that it will increase (the absolute value of the depolarizing field E_1 in this domain will decrease), while the polarization in domain 2 is negative, and its absolute value will decrease (the absolute value of the field E_2 will increase). We can summarize the result for polarization change in a compact form as

$$|P_2| < \tilde{P} < |P_1| < P_0, \tag{43}$$

$$|E_1| < |\tilde{E}| < |E_2|. \tag{44}$$

Now, using Eqs. (20) and (23), we can evaluate the pressure on the domain wall, when it is in an arbitrary position, as

$$p = \Phi_2 - \Phi_1 + \frac{\varepsilon_b}{2}(E_2^2 - E_1^2). \tag{45}$$

When the domain wall moves in the positive direction of the z axis ($z_1 > H/2$), it follows from Eq. (44) that E_2^2 increases and E_1^2 decreases, thus the last term in Eq. (45) increases. Thus, to show that the pressure increases when the wall is displaced from the central position, it is sufficient to check that the term $\Phi_2 - \Phi_1$ increases. Using Taylor expansion of polarization similar to Eqs. (29)–(31) and keeping only the linear terms in E_0, we find

$$\Phi_2 - \Phi_1 = A(\tilde{P}_1, \tilde{P}_2) + B(\tilde{P}_1, \tilde{P}_2)E_0, \tag{46}$$

where

$$A(\tilde{P}_1, \tilde{P}_2) = \tfrac{1}{4}\beta\left(\tilde{P}_1^2 - \tilde{P}_2^2\right)\left(2P_0^2 - \tilde{P}_2^2 - \tilde{P}_1^2\right), \tag{47}$$

$$B(\tilde{P}_1, \tilde{P}_2) = \frac{\tilde{P}_2\left(\tilde{P}_2^2 - P_0^2\right)}{3\tilde{P}_2^2 - P_0^2} - \frac{\tilde{P}_1\left(\tilde{P}_1^2 - P_0^2\right)}{3\tilde{P}_1^2 - P_0^2}. \tag{48}$$

Here, $\tilde{P}_1$, $\tilde{P}_2$, $\tilde{E}_1$, $\tilde{E}_2$ are polarizations and electric fields in nonsymmetric situation ($z_1 \neq H - z_1$) at zero external field in the domains 1 and 2, respectively.

In the case of zero external field, Eq. (43) leads to $\tilde{P}_2 < \tilde{P}_1 < P_0$, and it immediately follows that $A(\tilde{P}_1, \tilde{P}_2) > 0$. Thus, if the domain wall is displaced from the central position in the absence of external field, the direction of the force acting on the wall is parallel to the displacement. Thus, the equidistant periodic structure with charged domain walls is unstable. It corresponds to a maximum of the energy. In real systems, the position of charged domain walls can be stabilized because of the pinning by defects, as discussed above.

Now, we will analyze the second term in Eq. (46), which is proportional to E_0. Using Eqs. (48) and (38) we find

$$\frac{\partial B}{\partial z_1} \approx 6\left(\tilde{P}_1^2 - \tilde{P}_2^2\right)\frac{P_0^4\left(\tilde{P}_1^2 + \tilde{P}_2^2 - P_0^2\right) + 3P_0^2\tilde{P}_1^2\tilde{P}_2^2}{\left(3P_2^2 - P_0^2\right)^2\left(3P_1^2 - P_0^2\right)^2}\frac{\partial P}{\partial z_1}. \tag{49}$$

For the typical situation, where the depolarizing fields inside the domains are far from the thermodynamic coercive field, there is $\tilde{P}_1 \approx P_0$, $\tilde{P}_2 \approx P_0$. Thus, it follows from Eq. (49) using Eqs. (42) and (43) that it is $\partial B/\partial z_1 > 0$. Thus, when the domain wall is displaced from the central position by application of the external electric field, the pressure acting on the wall increases, and the criterion of successful poling is met. This means that the switching is governed by the condition in the periodic domain-wall pattern given by Eq. (36).

IV. CONCLUSIONS

A general formula for the local force acting on a domain wall with an arbitrary free charge on it has been obtained. In general, this force is not perpendicular to the domain wall. The force acting on the domain wall depends on the compensating free charge on it. This charge in turn depends on the characteristic size of the domain structure. A periodic domain structure with parallel head-to-head and tail-to-tail walls has been considered. It has been shown that a smaller domain size leads to a larger force. In a typical situation, the force acting on the charged domain wall is much smaller than the force acting on the neutral domain wall. Thus, poling of the sample with such a structure is difficult in comparison with poling of the sample with neutral domain walls. We also showed that in an ideal crystal, the periodic structure with charged domain walls is unstable. It corresponds to the maximum of the energy.

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APPENDIX A: EFFECTIVE STRESS TENSOR AND METHOD OF VIRTUAL DISPLACEMENT

The direct application of the formula for the generalized stress tensor, actually given in the book by Landau and Lifshitz[16] without derivation, leads to the force acting on the domain wall expressed as follows: $f_k = [[\Phi(P)]] - \hat{E}_i[[P_i]]n_k + \hat{E}_k\sigma_f + [[P_kE_i - P_iE_k]]n_i$. In comparison to the result, obtained using the method of virtual displacement, given by Eq. (10), this force contains an additional (in general nonzero) term $[[P_kE_i - P_iE_k]]n_i$. The contribution of this term at the pressure f_kn_k is equal to zero and in this case the result obtained with the method of virtual displacement is the same as that obtained using the stress tensor from the book by Landau and Lifshitz.[16]

APPENDIX B: DERIVATION OF THE LOCAL FORCE DENSITY ACTING ON THE ELEMENT OF THE DOMAIN WALL

We analyze the local pressure on the ferroelectric domain wall in a system shown in Fig. 4. We consider that inside a ferroelectric material with ferroelectric part of polarization $P_i^{(1)}$ there exists a closed domain of the material in a different polarization state $P_i^{(2)}$. The closed boundary splits the ferroelectric into two domains, which have volumes $V^{(1)}$ and $V^{(2)}$, respectively. Inside domains there are conductors, which carry charges $Q_E^{(1)}$ and $Q_E^{(2)}$ which are at electric potentials $\varphi_E^{(1)}$ and $\varphi_E^{(2)}$, respectively. We consider that the bound charges due to discontinuous change of polarization at the domain wall S_W are partially compensated by free charges of surface density σ_f. The charges on conductors and on the domain wall produce electric fields $E_i^{(1)}$ and $E_i^{(2)}$ within each domain. Symbols $\varphi^{(1)}$ and $\varphi^{(2)}$ stand for the electric potentials within each domain.

In order to obtain a general formula for the local pressure on domain wall, we follow the principle of virtual displacements, formulated in Sec. II. We will use the thermodynamic function G given by Eq. (8). In what follows, it is convenient to transform the work of electric sources into volume integrals

$$\varphi_E^{(1)}\,Q_E^{(1)} = -\varphi_E^{(1)}\int_{S_E^{(1)}} D_i^{(1)}n_i\,dS = \int_{V^{(1)}} E_i^{(1)}D_i^{(1)}\,dV + \int_{S_W}\varphi^{(1)}D_i^{(1)}n_i\,dS, \tag{B1a}$$

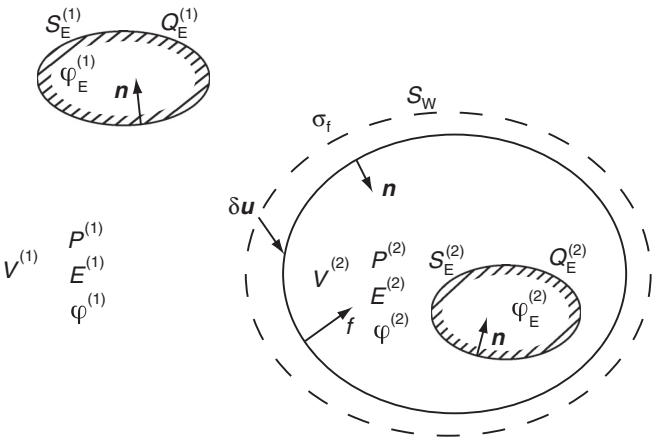

FIG. 4. General system of a ferroelectric in contact with two conductors. Domain wall S_W separates the ferroelectric into two domains of volumes $V^{(1)}$ and $V^{(2)}$. There are two conductors, which carry charges $Q_E^{(1)}$ and $Q_E^{(2)}$ and have electric potentials $\varphi_E^{(1)}$ and $\varphi_E^{(2)}$ within each domain. The quantities within each domain are the ferroelectric part of polarization $P_i^{(1)}$ and $P_i^{(2)}$, electric field $E_i^{(1)}$ and $E_i^{(2)}$, and electric potential $\varphi^{(1)}$ and $\varphi^{(2)}$. Symbol δu stands for the virtual displacement of the domain wall and symbol f stands for the local force density acting on a domain wall.

$$\varphi_E^{(2)}\, Q_E^{(2)} = -\varphi_E^{(2)} \int_{S_E^{(2)}} D_i^{(2)} n_i \, dS = \int_{V^{(2)}} E_i^{(2)} D_i^{(2)} \, dV - \int_{S_W} \varphi^{(2)} D_i^{(2)} n_i \, dS, \quad \text{(B1b)}$$

where $D_i^{(1)}$ and $D_i^{(2)}$ are the vectors of electric displacement in domains $V^{(1)}$ and $V^{(2)}$, respectively, and where we considered the absence of free charges, i.e., $\mathrm{div} D_i = 0$, inside domains $V^{(1)}$ and $V^{(2)}$ and vanishing the surface integral over the domain $V^{(1)}$ at infinity. Orientations of the normal vectors n_i are indicated in Fig. 4. Since the bulk quantities, e.g., φD_i, are generally discontinuous at the domain wall S_W, it will be useful to denote their jump by, e.g.,

$$[[\varphi D_i]] = \varphi^{(2)} D_i^{(2)} - \varphi^{(1)} D_i^{(1)}. \quad \text{(B2)}$$

If we employ the expression for electric displacement $D_i = P_i + \varepsilon_b E_i$, the thermodynamic function G can be expressed in a form

$$G = \int_{V^{(1)}} \left[\Phi^{(1)}(P^{(1)}) - \frac{1}{2}\varepsilon_b E_i^{(1)} E_i^{(1)} - E_i^{(1)} P_i^{(1)} \right] dV + \int_{V^{(2)}} \left[\Phi^{(2)}(P^{(2)}) - \frac{1}{2}\varepsilon_b E_i^{(2)} E_i^{(2)} - E_i^{(2)} P_i^{(2)} \right] dV + \int_{S_W} [[\varphi\,(P_i + \varepsilon_b\, E_i)]]\; n_i \, dS. \quad \text{(B3)}$$

Now, our task is to express the variation of thermodynamic function δG, which is produced by the virtual displacement of domain wall. When doing this, we have to keep in mind that the bulk quantities φ, E_i, and P_i are not independent variables. The relation between the electric field and the electric potential is given by $E_i = -\partial\varphi/\partial x_i$ and one has to consider the continuity of electric displacement at the domain wall, i.e., $[[P_i + \varepsilon_b\, E_i]]\; n_i = \sigma_f$. These additional conditions can be introduced into our treatment by adding new variables λ_i and μ according to the method of Lagrange multipliers. Using this approach, we obtain a new thermodynamic function $L = L(\varphi, E_i, P_i, \lambda_i, \mu)$, where *all bulk variables in the both domains are considered independent*:

$$L = \int_{V^{(1)}} \left[\Phi^{(1)}(P^{(1)}) - \frac{1}{2}\varepsilon_b E_i^{(1)} E_i^{(1)} - E_i^{(1)} P_i^{(1)} + \lambda_i^{(1)} \left(E_i^{(1)} + \frac{\partial\varphi^{(1)}}{\partial x_i} \right) \right] dV + \int_{V^{(2)}} \left[\Phi^{(2)}(P^{(2)}) - \frac{1}{2}\varepsilon_b E_i^{(2)} E_i^{(2)} - E_i^{(2)} P_i^{(2)} + \lambda_i^{(2)} \left(E_i^{(2)} + \frac{\partial\varphi^{(2)}}{\partial x_i} \right) \right] dV + \int_{S_W} \left[[[\varphi\,(P_i + \varepsilon_b\, E_i)]]\; n_i + \mu \left([[P_i + \varepsilon_b\, E_i]]\; n_i - \sigma_f \right) \right] dS. \quad \text{(B4)}$$

The virtual displacement δu of domain wall, which in general is not normal to the wall surface, shown in Fig. 4 produces the variation of all independent quantities $\delta\varphi$, δE_i, δP_i, $\delta\lambda_i$, and $\delta\mu$, except the electric potential on conductors $\varphi_E^{(1)}$ and $\varphi_E^{(2)}$, which are constant in the system. The variation of quantities results in the variation of thermodynamic function δL. In equilibrium, the variation δL equals the work produced by external force during the virtual displacement of domain wall

$$\delta L = -\int_{S_W} f_i \delta u_i \, dS, \quad \text{(B5)}$$

where f_i is the local force density per unit area of the domain wall, and S_W is the area of domain wall.

In order to express the variation δL, it is useful to consider the following formula for the variation of volume integral:

$$\delta \left\{ \int_V g \, dV \right\} = \int_V \delta g \, dV \pm \int_{S_W} g \, \delta u_k n_k \, dS, \quad \text{(B6a)}$$

where g is an arbitrary bulk quantity and plus and minus correspond to volume $V^{(1)}$ and $V^{(2)}$, respectively. The physical interpretation of the above formula is that the variation of volume integral includes the contribution due to variation of bulk quantity δg and the contribution due to the volume change of domains produced by the virtual displacement of domain wall δu. A similar formula can be written for the variation of surface integral over the domain wall:

$$\delta \left\{ \int_{S_W} g \, dS \right\} = \int_{S_W} \delta g \, dS + \int_{S_W} \frac{\partial g}{\partial x_k} \delta u_k \, dS, \quad \text{(B6b)}$$

where the first term on the right-hand side represents the contribution due to variation of bulk quantity δg on the domain wall and the second term represents the contribution due to change of the domain-wall position during the virtual displacement.

By applying Eqs. (B6) to function L given by Eq. (B4), we obtain

$$\delta L = \int_{V^{(1)}} \left\{ \delta P_i^{(1)} \left[\frac{\partial\Phi^{(1)}}{\partial P_i} - E_i^{(1)} \right] - \delta E_i^{(1)} \left[P_i^{(1)} + \varepsilon_b E_i^{(1)} - \lambda_i^{(1)} \right] + \frac{\partial\delta\varphi^{(1)}}{\partial x_i} \lambda_i^{(1)} + \delta\lambda_i^{(1)} \left[E_i^{(1)} + \frac{\partial\varphi^{(1)}}{\partial x_i} \right] \right\} dV + \int_{V^{(2)}} \left\{ \delta P_i^{(2)} \left[\frac{\partial\Phi^{(2)}}{\partial P_i} - E_i^{(2)} \right] - \delta E_i^{(2)} \left[P_i^{(2)} + \varepsilon_b E_i^{(2)} - \lambda_i^{(2)} \right] + \frac{\partial\delta\varphi^{(2)}}{\partial x_i} \lambda_i^{(2)} + \delta\lambda_i^{(2)} \left[E_i^{(2)} + \frac{\partial\varphi^{(2)}}{\partial x_i} \right] \right\} dV + \int_{S_W} \left\{ \delta\varphi^{(2)} \left[P_i^{(2)} + \varepsilon_b E_i^{(2)} \right] n_i - \delta\varphi^{(1)} \left[P_i^{(1)} + \varepsilon_b E_i^{(1)} \right] n_i + \left(\delta P_i^{(2)} + \varepsilon_b \delta E_i^{(2)} \right) n_i (\varphi^{(2)} + \mu) \right.$$

$$-\left(\delta P_i^{(1)}+\varepsilon_b\delta E_i^{(1)}\right)n_i(\varphi^{(1)}+\mu)+\delta\mu([[P_i+\varepsilon_b E_i]]n_i-\sigma_f)-\delta u_k\Bigg(\Bigg[\Bigg[\Phi(P)-\tfrac{1}{2}\varepsilon_b E_i E_i-E_i P_i$$
$$+\lambda_i\left(E_i+\frac{\partial\varphi}{\partial x_i}\right)\Bigg]\Bigg]n_k+\frac{\partial}{\partial x_k}([[\varphi(P_i+\varepsilon_b E_i)n_i]]+\mu([[P_i+\varepsilon_b E_i]]n_i-\sigma_f))\Bigg)\Bigg\}dS. \tag{B7}$$

The above formula can be further transformed using following integral expressions:

$$\int_{V^{(1)}}\lambda_i^{(1)}\left(\frac{\partial\delta\varphi^{(1)}}{\partial x_i}\right)dV+\int_{V^{(1)}}\left(\frac{\partial\lambda_i^{(1)}}{\partial x_i}\right)\delta\varphi^{(1)}dV=\int_{S_W}\lambda_i^{(1)}\delta\varphi^{(1)}n_i dS, \tag{B8a}$$

$$\int_{V^{(2)}}\lambda_i^{(2)}\left(\frac{\partial\delta\varphi^{(2)}}{\partial x_i}\right)dV+\int_{V^{(2)}}\left(\frac{\partial\lambda_i^{(2)}}{\partial x_i}\right)\delta\varphi^{(2)}dV=-\int_{S_W}\lambda_i^{(2)}\delta\varphi^{(2)}n_i dS, \tag{B8b}$$

where we consider that, during the virtual displacement of domain wall, the electric potential on conductors is kept constant by the electric sources, i.e. the variations of electric potentials on conductors are zero, and that the surface integral over domain $V^{(1)}$ is vanishing at infinity.

Applying Eqs. (B8) to formula (B7), we readily obtain the variation δL in a form

$$\delta L=\int_{V^{(1)}}\left\{\delta P_i^{(1)}\left[\frac{\partial\Phi^{(1)}}{\partial P_i}-E_i^{(1)}\right]-\delta E_i^{(1)}\left[P_i^{(1)}+\varepsilon_b E_i^{(1)}-\lambda_i^{(1)}\right]-\delta\varphi^{(1)}\frac{\partial\lambda_i^{(1)}}{\partial x_i}+\delta\lambda_i^{(1)}\left[E_i^{(1)}+\frac{\partial\varphi^{(1)}}{\partial x_i}\right]\right\}dV$$
$$+\int_{V^{(2)}}\left\{\delta P_i^{(2)}\left[\frac{\partial\Phi^{(2)}}{\partial P_i}-E_i^{(2)}\right]-\delta E_i^{(2)}\left[P_i^{(2)}+\varepsilon_b E_i^{(2)}-\lambda_i^{(2)}\right]-\delta\varphi^{(2)}\frac{\partial\lambda_i^{(2)}}{\partial x_i}+\delta\lambda_i^{(2)}\left[E_i^{(2)}+\frac{\partial\varphi^{(2)}}{\partial x_i}\right]\right\}dV$$
$$+\int_{S_W}\left\{\delta\varphi^{(2)}\left[P_i^{(2)}+\varepsilon_b E_i^{(2)}-\lambda_i^{(2)}\right]n_i-\delta\varphi^{(1)}\left[P_i^{(1)}+\varepsilon_b E_i^{(1)}-\lambda_i^{(1)}\right]n_i+\left(\varepsilon_b\delta E_i^{(2)}+\delta P_i^{(2)}\right)n_i(\varphi^{(2)}+\mu)\right.$$
$$-\left(\varepsilon_b\delta E_i^{(1)}+\delta P_i^{(1)}\right)n_i\left(\varphi^{(1)}+\mu\right)+\delta\mu([[P_i+\varepsilon_b E_i]]n_i-\sigma_f)-\delta u_k\Bigg(\Bigg[\Bigg[\Phi(P)-\frac{1}{2}\varepsilon_b E_i E_i-E_i P_i+\lambda_i\left(E_i+\frac{\partial\varphi}{\partial x_i}\right)\Bigg]\Bigg]n_k$$
$$\left.+\frac{\partial}{\partial x_k}([[\varphi(P_i+\varepsilon_b E_i)n_i]]+\mu([[P_i+\varepsilon_b E_i]]n_i-\sigma_f))\Bigg)\right\}dS. \tag{B9}$$

Employing the principle of virtual displacements, (B5) yields the formula for the local force density on the domain wall

$$f_k=\Bigg[\Bigg[\Phi(P)-\frac{1}{2}\varepsilon_b E_i E_i-E_i P_i+\lambda_i\left(E_i+\frac{\partial\varphi}{\partial x_i}\right)\Bigg]\Bigg]n_k-\frac{\partial}{\partial x_k}\{[[\varphi(P_i+\varepsilon_b E_i)n_i]]+\mu([[P_i+\varepsilon_b E_i]]n_i-\sigma_f)\}, \tag{B10}$$

equations of motion

$$\frac{\partial\Phi}{\partial P_i}=E_i, \tag{B11}$$

electric displacement $\lambda_i=\varepsilon_b E_i+P_i$, Gauss' law for electric displacement $\partial\lambda_i/\partial x_i=0$, relationship between electric field and electric potential $E_i=-\partial\varphi/\partial x_i$, continuity of electric potential at the domain wall $\mu=\varphi^{(1)}=\varphi^{(2)}$, and continuity of electric displacement at the domain wall $[[P_i+\varepsilon_b E_i]]n_i=\sigma_f$. Combining the above expressions, the formula for the local force density on the domain wall can be written in a form

$$f_k=\Bigg[\Bigg[\Phi(P)-\frac{1}{2}\varepsilon_b E_i E_i-E_i P_i\Bigg]\Bigg]n_k+[[E_k(P_i+\varepsilon_b E_i)]]n_i. \tag{B12}$$

Considering the continuity of tangential components of electric field at the domain wall $[[E_{t,i}]]=[[E_i-(E_k n_k)n_i]]=0$ and using the algebraic identity

$$[[fg]]=\widehat{f}\,[[g]]+[[f]]\,\widehat{g}, \tag{B13}$$

where $\widehat{f}=(f^{(1)}+f^{(2)})/2$ is the average of bulk quantity at the opposite sides of the domain wall, the general formula for the local force density of external sources on the domain wall can be further simplified to the form

$$f_k=[[\Phi(P)]]n_k-\widehat{E}_i\,[[P_i]]n_k+\widehat{E}_k\sigma_f. \tag{B14}$$

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