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## **Domain Structure in Ferroelectric Thin Films**

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### **Authors' contributions**

*This work was carried out in collaboration between both authors. Author AMD designed the study, supervised the work and proof read the draft before submission. Author NBY managed the literature searches, did the analyses under the the direction of AMD wrote the first draft of the manuscript. Both authors read and approved the final manuscript.*

### **Article Information**

DOI: 10.9734/AJR2P/2022/v6i230182

### **Open Peer Review History:**

This journal follows the Advanced Open Peer Review policy. Identity of the Reviewers, Editor(s) and additional Reviewers, peer review comments, different versions of the manuscript, comments of the editors, etc are available here: <https://www.sdiarticle5.com/review-history/89277>

**Received 06 May 2022**

**Accepted 13 July 2022**

**Published 19 July 2022**

**Original Research Article**

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## **ABSTRACT**

At low temperatures, uniaxial perovskites exhibit a spontaneous polarization with a permanent dipole moment. The broken symmetry state involves an order parameter and dielectric properties of the materials which are strongly dominated by a one-dimensional physics. For the last sixty years there has been growing interest in these crystals for potential applications in communication technology. Very recently, renewed interest to ferroelectric memory devices has been remarkable with particular emphasis on miniaturized devices. This new perspective comes along with great challenges, one of which is the critical size for stable domains in thin ferroelectric crystals. Since the polarization switching involves a pre-existing spontaneous polarization, it is of fundamental importance to address the question of conditions under which polarized domains can develop in a ferroelectric thin film. From previous articles, it has been observed that most studies focus on numerical simulations which is a good approach but for the fact that numerical simulations involve approximation of physical quantities, which is limitation to comparing experimental with theoretical studies. This motivated the authors to look purely at an analytical approach taking advantage of the new mathematical approach in the study of nonlinear systems.

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In this work, the authors have considered the question from an analytical point of view, focusing on an interesting model introduced by Lü and Cao. We propose an analytical counterpart of the numerical simulations done in this previous study.

*Keywords: Ferroelectric; thin film; polarization; switching; perovskites; analytical.*

## 1 INTRODUCTION

Ferroelectric materials exhibit a spontaneous polarization (i.e. are ordered) below a critical temperature  $T = T_c$  [1] known as Curie Temperature [1, 2]. The broken symmetry state involves an order parameter and dielectric properties of the materials which are strongly dominated by a one-dimensional physics [3, 4]. Indeed, due to their high dielectric constant, ferroelectrics are attractive candidates in designs of Dynamic Random Access Memory (DRAM) devices while the possibility to switch the polarization with an applied field can be utilized in cryptography and other data-security softwares. Very recently, renewed interest to ferroelectric memory devices has been remarkable with particular emphasis on miniaturized devices [5, 6, 7, 8, 9, 10, 11]. This new perspective comes along with great challenges, one of which is the critical size for stable domains in thin ferroelectric crystals.

Above  $T_c$ , the crystal lattice structure is macroscopically neutral or in its paraelectric phase [12]. In general, the spontaneous polarization in the ferroelectric phase can be reversed by the application of an electric field. Induced ferroelectricity is triggered either by mechanical stress or heat. The first factor is known as piezoelectric effect, and the second as pyroelectric effect [1, 13]. In crystals, elastic interactions between atoms can be modified by pressure or mechanical stress which affect the cohesion of the crystal lattice thus inducing a piezoelectric effect. For about two decades, the physics of ferroelectric materials has attracted a steady attention from both the experimental [1, 14, 18, 15, 16] and theoretical points of view [19, 17]. The earliest theory of ferroelectricity is the so-called Mean-Field approach based on the Ginzburg-Landau functional in which an order parameter  $P$  was introduced to describe the polarization [1, 15, 16, 20, 21]. Being a quartic polynomial in the order parameter, the Ginzburg-

Landau functional involves three extrema [22] of which a trivial solution  $P = 0$  describing the zero polarization state and two nonzero solutions  $P = P_{1,2}$  which in the absence of an applied electric field or mechanical stress become degenerate so that  $P_1 = -P_2$  or vice-versa. These two nonzero values of the polarization  $P$  are the two equilibrium positions between which the electric polarization can switch via a polarization reversal mechanism. The switching between the two degenerate polarization states can also be induced by an applied electric field resulting in a hysteresis loop.

In real contexts, the coupling between atoms carrying the polarizable charges creates a dispersion and promotes a displacement of the polarized electric charges. These phenomena, which reflect the lattice structure of the crystal, are described within the framework of the Ginzburg-Landau theory by a distortion energy [22] and the Ginzburg-Landau functional then turns to a Hamiltonian. In connection with this Hamiltonian form, a theory for the polarization dynamics in ferroelectric crystals has been constructed and explored at length with the help of the Hamiltonian formalism. Thus, it is now well established that the domain wall spanned by the polarization upon switching between its two equilibrium positions can be approximated by the so-called  $\phi^4$  kink solitary wave [22, 20, 21]. In the infinite-length limit, that is when the size of the ferroelectric device is very large compared to typical lattice parameter, the kink domain wall is a single smoothed-out function which is zero at  $x = 0$  but sharpens as  $x \rightarrow \pm\infty$ , where the polarization takes the values  $P = \pm P_o$ . Kinks and topological solitons in general, are robust against perturbations and various structural defects, a feature that makes them promising candidates in theoretical descriptions of the high stability of polarized domain structures observed in real ferroelectric devices.

Starting with a Ginzburg-Landau-Devonshire free energy widely used [23] in structural changes of imperfect crystals, Lü and Cao [19] investigated surface effects on profile, dynamics and stability of domain walls in embedded thin films. In their work, an approximation was made, namely, that the domain-wall shape does not change qualitatively and thus remains the same as the one known for infinite-length material, except that the amplitude and width of domains must now be determined by appropriate boundary conditions. In other words, they solve the equation governing the polarization dynamics in the infinite-length limit and next, apply finite-boundary conditions to extract the domain size and amplitude. From the viewpoint of fundamental physics and in view of recent progress in mathematical techniques for nonlinear equations, the above assumption of Lü and Cao [19] is rather questionable as it is now possible to directly solve nonlinear equations admitting soliton solutions, including the  $\phi^4$  equation which is also known as the classical Ginzburg-Landau equation, by quadrature.

The main goal of this paper is to exploit these recent mathematical approaches to improve numerical results of Lü and Cao [19] as concerns physical parameters of the domain wall in finite-size ferroelectrics.

The domain-wall solution will be derived in the form of the well-known  $\phi^4$  single-solitary-wave solution and following the classic paper of Krumhansl and Schrieffer [22]. We will first look at the Landau free energy next authors look at model of Lü and Cao [19] model using a more physical considerations which allows the obtaining of exact results by a direct quadrature method. The last section will be devoted to conclusion and perspectives.

## 2 DOMAIN WALLS IN FERROELECTRIC THIN FILMS

Considering the model for domain-wall structures in ferroelectric thin films proposed by Lü and Cao [19], in which they used numerical simulations to provide physical parameters are given particular values. This has a drawback since it does not permit consistent quantitative and qualitative comparisons of theoretical results

with experimental findings. After discussing their numerical results, authors will consider an integration method which leads to an exact analytical kink solution for the  $\phi^4$  equation and which reflects the finite size of the thin film.

Their model [19] consist of a thin ferroelectric film of finite thickness i.e.  $-L_{1s} < x < L_{2s}$ , embedded in two thin surface layers each surrounded by an electrode. The easy polar axis of the film is assumed to be normal to the film surface and the film is in a single domain state. Starting with the Ginzburg-Landau-Devonshire energy for the system 1:

$$G_L = G_o + \int_{-L}^L dx \left\{ \frac{1}{2} A [T - T_b \psi(x)] P^2 + \frac{1}{4} C P^4 + \frac{1}{6} D P^6 + \frac{1}{2} K \left( \frac{dP}{dx} \right)^2 - \frac{1}{2} E_d P - \bar{E}_{ie} P - EP \right\}. \quad (1)$$

with  $A$ ,  $C$ ,  $D$  and  $K$  are independent of temperature  $T$  and position  $x$ . For a first-order phase transition  $A$ ,  $D$  and  $K$  must be positive and  $C$  negative. For a second-order phase transition,  $A$ ,  $C$  and  $K$  must be positive and  $D = 0$ .  $T_b$  is the transition temperature of bulk material,  $E$  is an applied external field which is uniform along the  $x$  direction. The direction of average effective internal bias field  $\bar{E}_{ie}$  is parallel to the direction of the easy polarization of an asymmetric ferroelectric film,  $\bar{E}_{ie} = 0$  if there is inversion symmetry. Lastly,  $E_d$  is the depolarization field defined as:

$$E_d = -\frac{1}{\epsilon_o} (P - \bar{P}) \quad (2)$$

where  $\epsilon_o$  is the vacuum dielectric permittivity. We define the average polarization  $\bar{P}$  as:

$$\bar{P} = \frac{1}{2L} \int_{-L}^L P(x) dx. \quad (3)$$

In general, the depolarization field is irrelevant if the system is perfect up to the surface and the surfaces are coated with metal electrodes. It is also irrelevant if there are injected charges that neutralize completely the bound surface charges as well established [24]. The function  $\psi(x)$  in formula (1) represents the inhomogeneous nature of the surface layer. To ensure the

continuity of the polarization  $P(x)$  and its derivative in the whole region of interest, their require that:

$$\psi(-L_{1s}) = \psi(L_{2s}) = 1, \quad \text{and} \quad \frac{d\psi}{dx} \Big|_{x=-L_{1s}} = \frac{d\psi}{dx} \Big|_{x=L_{2s}} = 0, \quad (4)$$

The Hamilton-Jacobi equation obtained from the Ginzburg-Landau-Devonshire energy (1) is:

$$K \frac{d^2 P}{dx^2} = A [T - T_b \psi(x)] P + C P^3 + D P^5 - E_d - \bar{E}_{ie} - E. \quad (5)$$

Lü and Cao solved equation (5) numerically in the structural regime, i.e. for ferroelectric instabilities of the second order. In this regime, the crystal potential is of a double-well shape and the parameter  $D = 0$ . The key point in Lü-Cao's numerical analysis is the assumption of the boundary condition:

$$\frac{d\psi}{dx} \Big|_{x=\pm L} = 0. \quad (6)$$

This boundary condition means that the polarization field within a domain extends in the space up to the surface layers and has a vanishing shape in the electrodes. From a physical point of view, authors can understand this boundary condition as follows: because of the presence of the electrodes, bound charges are completely neutralized by free charges on the electrode surfaces. Consequently no charge coming from the thin film is lost, but is compensated for by the electrodes which is a useful constraint to guarantee loss or non volatility of the ferroelectric memory.

With the above boundary conditions, the authors solved equation (5) in the continuum regime with  $D = 0$  and considering the following forms for the quantity  $\psi(x)$ :

$$\psi(x) = \begin{cases} 1 - \left(\frac{x+L_{1s}}{\lambda_1}\right)^2, & -L \leq x \leq -L_{1s} \\ 1, & -L_{1s} \leq x \leq -L_{2s}, \\ 1 - \left(\frac{x-L_{2s}}{\lambda_2}\right)^2, & -L_{2s} \leq x \leq -L. \end{cases} \quad (7)$$

Since their model combines different materials, it can readily be treated as a perfect material in which each layer, which represents a distinct material, is identified by a proper parameter. In equation(7) it is the degree of imperfection  $\lambda_i$  that characterizes the materials.

Lü-Cao's numerical simulations were carried out assuming two distinct cases, first they kept the thickness of each of the three layers fixed and vary their degrees of imperfection, and next vary the layers but fix the degrees of imperfection.

Comparing their results with some experimental results [25], authors observe full shape of domain-wall patterns which, that form a periodic structure implying a dominant periodic ordering of domain walls in polarized domains of the thin film. We also observed the shape of a single domain wall in the periodic domain; it suggests that "kink" profile displayed by the single domain wall of the infinite-length system is also present. However, because of the periodic ordering, domain walls are nucleated and hence loose their full kink shape. On the otherhand, Lü and Cao numerical simulations show a twinned kink or kink-antikink patterns which are consistent with the full shape of domain-wall patterns.

### 3 THE GOVERNING EQUATION

In this section authors have developed an analytical method for solving the equation governing the shape and dynamics of domain walls in thin ferroelectric films taking into consideration the model proposed by Lü and Cao [19].

The motivation of an analytical solution to the problem is the need for a general solution that permits more effective comparison of theoretical predictions with experimental results over a broad range of parameter values, contrary to numerical simulations where all physical parameters are given arbitrary numerical values.

In the displacive regime, the parameter  $D = 0$  and equation (5) which governs the spatial shape profile of domain walls in the presence of the external field reduces to:

$$K \frac{d^2 P}{dx^2} = A [T - T_b \psi(x)] P + C P^3 - E_d - \bar{E}_{ie} - E. \quad (8)$$

To also take into account the dynamics of domain wall, it is useful to rewrite equation (8) as a nonlinear partial differential equation:

$$M P_{tt} - K P_{xx} - A [T - T_b \psi(x)] P - C P^3 + E_d + \bar{E}_{ie} + E = 0. \quad (9)$$

where  $M$  is the effective mass of the polarized atom. Defining new parameters as:

$$f = \frac{P}{P_o}, \quad P_o = P_1, \quad e_{ie} = \frac{\epsilon_0 E_{ie}}{P_o}, \quad e = \frac{\epsilon_0 E}{P_o}, \quad \bar{f} = \bar{P}/P_o, \\ a = \tau - \psi(x), \quad \tau = T/T_b, \quad \sigma = \frac{1}{\epsilon_o A T_b}, \\ F = (e_{ie} + e + \bar{f}) \epsilon_o / P_o. \quad (10)$$

and introducing a new variable  $z = x - \vartheta t$  where  $\vartheta$  is the domain-wall velocity, equation (10) reduces to:

$$f_{zz} - \frac{1}{\ell^2} [a f - f^3 - \sigma (f - F)] = 0, \quad (11)$$

with

$$\ell^2 = \frac{M c_o^2}{A T_b \gamma^2}, \quad \gamma^2 = 1 - \vartheta^2 / c_o^2, \quad (12)$$

where  $\gamma$  is the Galilean contraction factor. A further simplification of equation (11) yields:

$$f_{\zeta\zeta} - a f - f^3 - \sigma (f - F) = 0, \quad \zeta = z/\ell. \quad (13)$$

This last equation is the dimensionless  $\phi^4$  equation with an additional term accounting for an applied constant external field, of effective magnitude  $\sigma F$ . It is interesting to note that, the actual shape of the domain wall describing the spontaneous polarization as the temperature is lowered below the Curie temperature  $T_b$  of the embedded ferroelectric film, is determined by the homogeneous  $\phi^4$  equation since this is the exact equation provided by the double-well potential of the crystal lattice in this temperature regime. Thus, the external field is applied only after the high-temperature crystal symmetry has been broken towards the ferroelectric phase characterized by the polarization order parameter. Therefore, the shape of the domain wall in the absence of applied field is more relevant and will be discussed separately.

### 3.1 Domain-wall Solutions in the Absence of an External Field

In the absence of the external field, (13) turns to the  $\phi^4$  equation which the authors will solve in this section. Multiplying (13) (in which authors have set  $F = 0$ ) by  $f_\zeta$  and integrating once with respect to  $\zeta$ , authors find:

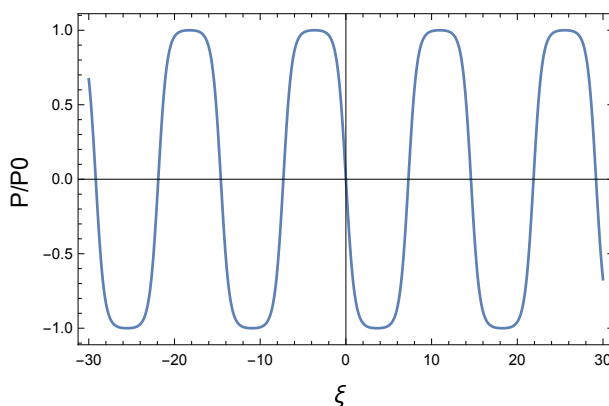
$$f_\zeta^2 = \frac{a}{2} (\alpha^2 - f^2) (\beta^2 - f^2), \quad (14)$$

where  $\alpha$  and  $\beta$  are arbitrary parameter connected to the constant of integration. By grouping terms

which are functions of the variables  $\zeta$  and  $f$  separately and arrive at the following integral equation:

$$\int \frac{df}{\sqrt{(\alpha^2 - f^2)(\beta^2 - f^2)}} = \sqrt{\frac{a}{2}} \int d\zeta. \quad (15)$$

Equation (15) is a member of the elliptic integral equations [26, 27] whose solutions are in general given in terms of Jacobi Elliptic functions. In this specific case, this equation is solved for a finite-length system that is, with the boundary



**Fig. 1. Periodic domain wall solution of the  $\phi^4$  equation**

conditions:

$$f \rightarrow f_{1,2} \quad \text{when} \quad \zeta \rightarrow \pm L, \quad (16)$$

where the total size of the system is assumed to be  $2L$  and  $f_{1,2}$  are defined with respect to the two equilibrium values of the polarization already defined. In addition, it is also require that:

$$\frac{df}{d\zeta} \Big|_{z=\pm L} = 0, \quad (17)$$

which expresses the jump of the polarization field on crossing the interface between the thin film and the surface layer.

From a general standpoint, equation (15) can be solved irrespective of the boundary conditions (16) and (17). However, once the general solutions are obtained, the defined boundary conditions must be used as specific constraints to evaluate arbitrary parameters such as the amplitude, periods and width understanding that authors expect nonlinear periodic kink structures.

In terms of Jacobi Elliptic functions [20, 21, 27] the integral equation (15) leads to the solution:

$$f(\zeta) = f_{1,2} \operatorname{sn} \sqrt{\frac{2a}{1+\kappa^2}} \frac{\zeta - \zeta_0}{\sqrt{2}}, \quad (18)$$

$$f_{1,2} = \pm \sqrt{\frac{-2a\kappa^2}{1+\kappa^2}},$$

where  $\operatorname{sn}$  is more precisely the Jacobi snoidal function and  $\kappa = \alpha/\beta$  is its modulus. As Fig. 1. indicates, the last solution describes a periodic structure made of ordered kink-antikink solitary waves of the  $\phi^4$  type. Associated with this periodic feature is the period  $L_\kappa$ , which is a characteristic parameter of the snoidal function  $\operatorname{sn}$  determined by the condition:

$$f(\zeta + L_\kappa) = f(\zeta), \quad (19)$$

and which reads:

$$L_\kappa = \kappa K(\kappa), \quad (20)$$

where  $K(\kappa)$  is the Elliptic integral of the first order. Interestingly enough, in connection with the inherent periodicity of the domain-wall solution obtained in (18) and which emerges in Fig. 1, the authors can postulate that the constraint of a finite size favours periodic structures. In fact this postulate is well conformed by the experiment [25] as discussed early. Before closing this section, it is also useful to mention the relevant fact that the periodic arrangement of kink domain walls has to accommodate the size  $L$  of the system to guarantee their stability. Accordingly, the size of the thin film should be proportional to the period  $L_\kappa$ . This last remark clearly shows that Jacobi Elliptic functions offer a great advantage in their use for describing nonlinear excitations in finite-size systems, as done in many other physical contexts.

### 3.2 Domain-wall Solutions in the Presence of an External Field

Returning to equation (13), a solution describing the effect of an applied field on the domain wall structure in the finite-size system is seen. Once again, multiplying (13) by  $f_\zeta$  and integrate once with respect to  $\zeta$ . The authors obtain:

$$f_\zeta^2 = 2 \left( \frac{f^4}{4} + a_o \frac{f^2}{2} - b f + k \right) \quad (21)$$

with  $k$  the constant of integration,  $a_o = a + \sigma$  and  $b = \sigma F$ . The right hand side of (21) is a quartic polynomial in  $f$ , in general it can also be rewritten in factorized form as:

$$F(f) = (f - \alpha)(f - \beta)(f - \gamma)(f - \xi) \quad (22)$$

where  $\alpha, \beta, \gamma$  and  $\xi$  are the four roots of the quartic polynomial. With this factorized expression, (21) can now be transformed into the integral equation:

$$\int_0^u \frac{df}{\sqrt{(f - \alpha)(f - \beta)(f - \gamma)(f - \xi)}} = \sqrt{\frac{1}{2}} \int d\zeta. \quad (23)$$

According to the table of integrals [26], the last integral equation involves several distinct kinds of solutions in relation to all the possible combinations between the four roots. However, to reduce the number of possible combinations, the authors take the case without the external field treated previously as a reference, and choose the roots in such a way that the problem reduces to this reference when  $F = 0$ . With this assumption, the authors obtain the combination  $u > \alpha > \beta > \gamma > \xi$  as more appropriate for this context. The table of integrals then yields:

$$\frac{2}{\sqrt{(\alpha - \gamma)(\beta - \xi)}} F(\phi, q_o) = \sqrt{\frac{1}{2}} \zeta \quad (24)$$

where  $F(\phi, q_o)$  is the Jacobi Elliptic integral of the first kind,

$$\begin{aligned} \phi &= \arcsin \sqrt{\frac{(\beta - \xi)(u - \alpha)}{(\alpha - \xi)(u - \beta)}}, \\ q_o &= \sqrt{\frac{(\beta - \gamma)(\alpha - \xi)}{(\alpha - \gamma)(\beta - \xi)}}. \end{aligned} \quad (25)$$

To extract the variable  $u$  hidden in the argument of the Elliptic integral  $F(\phi, q_o)$ , authors use a trick which consists of introducing Jacobi Elliptic functions via the twelve Jacobi identities, which in our particular case allows us setting:

$$\sin \nu = sn u. \quad (26)$$

Otherwise, remarking that

$$\sin \phi = \sqrt{\frac{(\beta - \xi)(u - \alpha)}{(\alpha - \xi)(u - \beta)}} \quad (27)$$

which follows from (25), authors find:

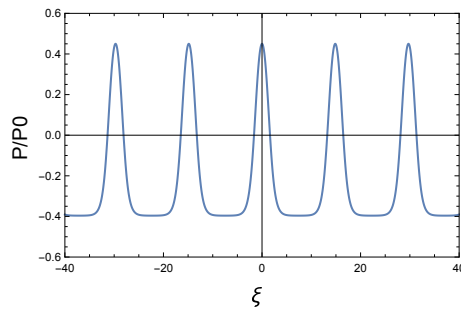
$$\begin{aligned} \frac{(\beta - \xi)(u - \alpha)}{(\alpha - \xi)(u - \beta)} &= sn^2 B\zeta, \\ B &= \frac{\sqrt{(\alpha - \gamma)(\beta - \gamma)}}{2} \end{aligned} \quad (28)$$

and from the last relation authors derive  $f \equiv u$ ;

$$f(\zeta) = \frac{\alpha(1 - \frac{\beta}{\alpha\lambda} sn^2 B\zeta)}{(1 - \frac{1}{\lambda} sn^2 B\zeta)}, \quad \lambda = \frac{\beta - \xi}{\alpha - \xi}. \quad (29)$$

As it stands, the last solution is rather cumbersome and does not allow at all a simple interpretation. In addition, although authors imposed  $\alpha > \beta > \gamma > \xi$  among the four roots of the quartic polynomial  $F(f)$ , these roots can be explicitly determined with the help of quartic factorization tools in Mathematica software. For this last purpose, authors expand the factorized form of  $F(f)$ , compare coefficients and solve the resulting  $4 \times 4$  matrix equation. Nevertheless, it is quite easy to check that when  $F = 0$ , the quartic polynomial  $F(f)$  involves only even powers in  $f$ . Therefore the four roots become doubly degenerate and fall into two sets of two mutually conjugate roots which is exactly what authors had in (14). The Fig.2. shows that the domain walls are still periodic and persistent.

The kink solution of the  $\phi^4$  equation with an external bias has already been found in the infinite-length limit [21, 28, 29, 30, 31]. The work [30] suggest that for a relatively weak applied field the kink shape is not affected but instead, is accelerated if its motion is shifted from its zero-field position by a finite spatial amount, equivalent to the effect of a "Goldstone translation". However, if the magnitude of the field is large enough the field can result into a deformation of the kink shape as shown in Fig. 2, thus leading to an asymmetric kink.



**Fig. 2. Periodic domain wall in the presence of an external field**

In fact, the formation of asymmetric kink is not dramatic to the stability of the system, instead it reflects the capability of the domain wall to adapt itself to conditions of its medium imposed by external factors. Basing on these remarks, authors can readily expect the nonlinear solution obtained in (29) to stand as a periodic counterpart of the asymmetric single-kink solution obtained for the  $\phi^4$  equation with an external bias [28, 29, 30, 31].

## 4 CONCLUSION

Authors have investigated the structure of domains and domain walls in the ferroelectric phase of perovskite crystals with finite sizes and show that, these systems are promising candidates for the design of non-volatile thin ferroelectric Random Access Memory devices. Authors considered a specific model introduced by Lü and Cao whose numerical results are in good agreement with experimental predictions for single domain, and have proposed an analytic theory in support of these numerical results. In this particular case when the external field is switched off, authors found an analytical soliton solution whose periodic feature is also in good agreement with both experiments and simulations of Lü and Cao. In the case when the applied field is not zero, even though the analytical solution is rather cumbersome and does not permit simple interpretation with respect to available experimental and numerical results, nevertheless, authors have shown that this complicated general solution has the homogeneous solution which is still periodic and reduces to the well known asymmetric kink

solution of the infinite-length  $\phi^4$  model with external field. Thus, despite an apparent complex form the general solution proposed for the first time in this work is relevant.

## ACKNOWLEDGEMENT

The Authors wish to thank The Laboratory of Research on Advanced Materials and Nonlinear Sciences (LaRAMaNS) is partially funded by the World Academy of Sciences (TWAS), Trieste, Italy.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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