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An approach to the Klein–Gordon equation for a dynamic study in ferroelectric materials

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Abstract

Ferroelectric materials such as lithium niobate and lithium tantalate show a non-linear hysteresis behaviour, which may be explained by dynamical system analysis. The behaviour of these ferroelectrics is usually explained by domains and domain wall movements. So, the spatial variation of the domain wall was studied previously in order to see its effect on the domain wall width in the context of the Landau–Ginzburg functional. In the present work, both temporal and spatial variations of polarization are considered, and by using the Euler–Lagrange dynamical equation of motion, a Klein–Gordon equation is derived by taking the ferroelectrics as a Hamiltonian system. An interaction has been considered between the nearest neighbour domains, which are stacked sideways in a parallel array with uniform polarization. This interaction term is associated with the spatial term and when this interaction is assumed to be zero, the spatial term vanishes, giving rise to a Duffing oscillator differential equation, which can be also studied by a dynamic system analysis.

1. Introduction

Important properties such as giant polarization, and many other properties, are commonly observed in ferroelectric materials, which have a wide area of applications [1]. However, the most significant property is their ability to show non-volatile memory, which remains finite when the field is withdrawn ([2] and the other important references therein). This is clearly noticed in the hysteresis curve of polarization (P) versus external electric field (E), which is non-linear. The study of this non-linear behaviour of P versus E is quite interesting in order to get an insight into the memory function. In a previous investigation, we used the famous Landau–Ginzburg functional for the free energy (G) to explain this behaviour [3].

This was done by giving perturbations on P , E and the Landau free energy (G) by taking the spatial variation of the domain wall width in terms of these parameters as ordinary differential equations. By treating them as an eigenvalue problem, the critical values of polarization (P_c) were estimated within the ‘zone of stability’ through a linear Jacobian transformation, which showed a possibility of a ‘giant memory’, and the corresponding limits of domain wall width were also found in the case of lithium tantalate and lithium niobate ferroelectric crystals [3], whose switching and hysteresis behaviour have been studied in detail by Gopalan *et al* [4].

In the previous study, only spatial variation of polarization was considered through a static soliton wave solution of Lines and Glass [5] as a governing equation, as was also done by Kim *et al* [2]. It is important to study polarization as a function of time for a better understanding of the memory function and consequent switching phenomenon, which leads to a dynamical situation. Moreover, it is also important to study the variation of both space and time of polarization with the Euler–Lagrange equation of motion involving an ‘interaction’ between the neighbouring domains associated with the spatial term, which gives rise to a ‘field equation’ like the Klein–Gordon equation. This Klein–Gordon (KG) equation can of course be used for dynamic system analysis. However, if the interaction term tends to be very small for certain ferroelectric crystals so that it could be assumed to be zero, the spatial term vanishes and we get a Duffing oscillator non-linear differential equation. This equation is also important for dynamic system analysis for certain device applications in the time domain [6].

Such studies have not been undertaken so far, possibly due to an over-emphasis on the Landau–Ginzburg equation, which in our case is taken as the potential formulation in the context of dynamical systems. Here, it should be clearly mentioned that the depolarizing field term, domain wall energy and the elastic energy are excluded in this analysis in order to show that even the Landau potential is quite enough to explain the dynamic situation in ferroelectric materials. A detailed theoretical analysis is presented here in terms of constructing a Hamiltonian and then using a ‘variation principle’ to arrive at the Klein–Gordon equation. This classical approach seems quite powerful to enlighten us on the realm of physics involved in such interesting and important condensed matter systems with various applications.

2. Theoretical development

The relation of the free energy (G) to the order parameter (P) is described by the Landau–Ginzburg equation, neglecting the higher order terms, as

$$G = \left(-\frac{\alpha_1}{2} P^2 + \frac{\alpha_2}{4} P^4 \right) - EP. \quad (1)$$

Here, EP is the energy due to the applied electric field, and α_1 and α_2 are the expansion coefficients, which are both greater than zero and which are also important parameters for ferroelectric materials, as shown later.

2.1. Hamiltonian formulation

In the previous study, two domains in the form of rectangular boxes were taken in order to find the optimal width of the domain wall between these two domains on which the perturbation was given with respect to spatial coordinates [3]. In a Hamiltonian system such as the dipolar system, since we are interested in the evolution of both space and time of the polarization (P), we are inclined to take a series of N 2D rectangular boxes with uniform polarization as an ‘array of domains’, and then first of all study the time evolution of polarization of the i th domain (i.e. P_i). Now, let us write the Hamiltonian of such a system of domains in terms of the

momentum (p_i) in the kinetic part and consider equation (1) for the free energy functional in the potential formulation as

$$\mathbf{H} = \sum_{i=1}^N \left(\frac{1}{2m_d} \right) p_i^2 + \sum_{i=1}^N \left(\left(-\frac{\alpha_1}{2} P_i^2 + \frac{\alpha_2}{4} P_i^4 \right) - E P_i \right). \quad (2)$$

The momentum can be defined in terms of the order parameter (P_i) as

$$p_i = \frac{\partial}{\partial t} \left(\frac{m_d}{Q_d} P_i \right) = \left(\frac{m_d}{Q_d} \right) \dot{P}_i. \quad (3)$$

Here, we define m_d = mass per unit volume of dipoles (mass density of dipoles) and Q_d = charge per unit volume of dipoles (charge density of dipoles). The concept of mass and charge in a dipolar material is lucidly given by Todorov [7]. Equation (3) can be used to write the Hamiltonian in terms of the order parameter (P_i) as

$$\mathbf{H} = \sum_{i=1}^N \frac{m_d}{2Q_d^2} (\dot{P}_i)^2 + \sum_{i=1}^N \left(\left(-\frac{\alpha_1}{2} P_i^2 + \frac{\alpha_2}{4} P_i^4 \right) - E P_i \right). \quad (4)$$

This Hamiltonian gives rise to the Duffing oscillator differential equation for dynamic system analysis [6].

2.2. Interaction in a static field

In order to take a coupling or interaction between the neighbouring domains, i.e. between the polarizations in the i th domain (P_i) and the polarization in $(i - 1)$ th domain (P_{i-1}) and the polarization in the $(i + 1)$ th domain (P_{i+1}), it is useful to operate in the generalized coordinate system of (p_i, q_i) , where p_i is the momentum of the system, as in equation (3), and $q_i = P_i/Q_d$. Since the dimension of Q_d is Coulomb m^{-3} , the dimension of P_i/Q_d is only metres, and it is then useful to eventually introduce the ‘space’ as a variable.

This is an important point in that even without considering the depolarizing or domain wall energy, the space term can be introduced in the energy formulation in the construction of our Hamiltonian for the analysis of the equation of motion of this dipolar system. The elastic energy term is also important, but it can be considered to be renormalized into the expansion coefficients of equation (1). Hence, the Landau free energy is just sufficient for potential calculation. Now, we can write the Hamiltonian of this system, as in equation (4), by taking care of the above interaction terms, as

$$\begin{aligned} \mathbf{H} &= \frac{1}{2m_d} \sum_{i=1}^N p_i^2 + \sum_{i=1}^N \left[-\frac{\alpha_1 Q_d^2}{2} \left(\frac{P_i}{Q_d} \right)^2 + \frac{\alpha_2 Q_d^4}{4} \left(\frac{P_i}{Q_d} \right)^4 \right] \\ &\quad + \sum_{i=1}^N \left[\frac{k Q_d^2}{4} \left(\frac{P_i}{Q_d} - \frac{P_{i-1}}{Q_d} \right)^2 - Q_d \left(\frac{P_i}{Q_d} \right) E \right] \\ &= \frac{m_d}{2} \sum_{i=1}^N (\dot{q}_i)^2 + \sum_{i=1}^N \left[-\left(\frac{\alpha_1 Q_d^2}{2} \right) q_i^2 + \left(\frac{\alpha_2 Q_d^4}{4} \right) q_i^4 \right] \\ &\quad + \sum_{i=1}^N \left[\frac{k Q_d^2}{4} (q_i - q_{i-1})^2 - Q_d q_i E \right]. \end{aligned} \quad (5)$$

Here, $(k/2)(P_i - P_{i-1})$ is considered to be the contribution by first neighbour coupling or interaction. In a ferroelectric system, this kind of coupling can be considered simplistic, but surely for this type of arrangement of domains, there has to be some kind of near-neighbour coupling between the domains in one dimension, as mentioned above. The above Hamiltonian

involves an energy density in order to evaluate the Lagrangian density. By using the Euler-Lagrange equation of motion, we can write

$$m_d(\ddot{q}_i) = -[-(\alpha_1 Q_d^2)q_i + (\alpha_2 Q_d^4)q_i^3] - \frac{kQ_d^2}{2}(q_i - q_{i-1} + q_i - q_{i+1}) + Q_d E \quad (6)$$

$$\left(\frac{m_d}{Q_d^2}\right)\ddot{P}_i = \alpha_1 P_i - \alpha_2 P_i^3 + \frac{k}{2}(P_{i+1} - 2P_i + P_{i-1}) + E \quad (7)$$

$$\left(\frac{m_d}{Q_d^2}\right)\ddot{P}_i - \alpha_1 P_i + \alpha_2 P_i^3 = \frac{k}{2}(P_{i+1} - 2P_i + P_{i-1}) + E. \quad (8)$$

At this stage, it should be mentioned that in order to take both temporal and spatial variation of polarization, we have taken the time variation from the kinetic part, but the spatial variation comes out from the ‘interaction’ between the polarization domains, which are stacked sideways in the x -coordinate. In our derivation of equation (8), the spatial variation of polarization (P_i) is considered as discrete. Now, in order to change from a discrete to the continuum KG system, it is necessary to give some physical explanations for our system. The number of domains is N and let us consider the ‘domain length’ as L so that the sample dimension along the x -coordinate is NL , which is significantly larger than L .

Therefore, the change in polarization while moving from one domain to the next one occurs within a ‘length scale’ which is much smaller compared to that of the sample (NL). In this length scale, we can replace $P_i(t)$ by $P(x, t)$. Similarly, we can replace $P_{i-1}(t)$ by $P(x - L, t)$, and $P_{i+1}(t)$ by $P(x + L, t)$. Now, we can write the polarization terms arising from the interaction in equation (8) as

$$\begin{aligned} P_{i+1}(t) - 2P_i(t) + P_{i-1}(t) &= P(x + L, t) - 2P(x, t) + P(x - L, t) \\ &= L^2 \frac{\partial^2 P}{\partial x^2} + O(L^4) \end{aligned} \quad (8a)$$

after Taylor expansion of the above polarization terms⁴. Then, it is possible to describe this scenario as

$$\frac{P(x + L, t) - 2P(x, t) + P(x - L, t)}{L^2} \approx \frac{\partial^2 P}{\partial x^2} \quad (8b)$$

since L is very small, in the nanometre range. This is the key point in this particular derivation of the KG equation for our system of ferroelectrics. Now, by introducing equation (8b) into (8) and after dropping the index i , we get

$$\left(\frac{m_d}{Q_d^2}\right)\ddot{P} - \alpha_1 P + \alpha_2 P^3 - \left(\frac{k}{2}\right)L^2 \frac{\partial^2 P}{\partial x^2} - E = 0. \quad (9)$$

It may be mentioned that Rikvold *et al* [8] considered an oscillating field in the context of a dynamic phase transition by dividing the ‘space’ into lattice points with lattice spacing $\Delta x = a$, which was done for the purpose of numerical integration.

For the convenience of analysis as in the previous study [3], different participating variables along with the spatial term are taken in the non-dimensional form as

$$P' = \frac{P}{P_s} \quad (10a)$$

$$E' = \frac{E}{E_c} \quad (10b)$$

$$t' = \frac{t}{t_c} \quad (10c)$$

⁴ This important point was raised by the referee.

$$x' = \frac{x}{L} \quad (10d)$$

where P_s = saturation polarization, E_c = coercive field in the usual hysteresis curve, t_c = critical time, which is equal to the total time taken for the external field to reach a saturation value on the higher field side at $P = P_s$, and here, L is the length of the one-dimensional single domain in the spatial x -coordinate.

Equation (9) can now be written as

$$\left(\frac{m_d}{Q_d^2}\right)\left(\frac{P_s}{t_c^2}\right)\frac{\partial^2 P'}{\partial t'^2} - (\alpha_1 P_s)P' + (\alpha_2 P_s^3)P'^3 - \left(\frac{k P_s}{2}\right)\frac{\partial^2 P'}{\partial x'^2} - E_c E' = 0. \quad (11)$$

Now, let us put $t_c = \frac{1}{Q_d} \sqrt{\frac{m_d P_s}{E_c}}$ seconds, and for the sake of simplicity by removing the prime notation and by taking $\alpha_2 = \alpha_1 / P_s^2$ from [2], we get

$$\frac{\partial^2 P}{\partial t^2} - \bar{\alpha}_1 P + \bar{\alpha}_1 P^3 - \left(\frac{k P_s}{2 E_c}\right)\frac{\partial^2 P}{\partial x^2} - E = 0 \quad (12)$$

where $\bar{\alpha}_1 = \bar{\alpha}_2 = \frac{\alpha_1 P_s}{E_c}$. This is the famous Klein–Gordon (KG) equation. Therefore, if we take both time and space as variables, we arrive at the KG equation, which should be the ‘governing’ equation of polarization in terms of the Euler–Lagrange equation of motion for charged particles (dipoles) in a ferroelectric material consisting of an ‘array of domains’ with coupling/interaction between the neighbouring domains.

The KG equation is actually a classical ‘field’ equation, which is applicable to electric charges or dipoles with two energy eigenvalues describing two different charge states. It is pertinent to mention here that the soliton solution of the KG-like equation is treated as a structureless ‘point-like’ particle, and the topological solitons possess an important application to describe domain walls in ferroelectric materials, as shown by Gonzales *et al* without mentioning any particular ferroelectric system [9], whereas we have actually done the stability analysis in the case of lithium niobate and lithium tantalate [10].

3. Results and discussion

In the above KG equation (12), if we take the interaction constant $k = 0$ as a special case for certain ferroelectric crystals where the interaction between the neighbouring domains is weak, i.e. the k term is very small, then we arrive at the Duffing oscillator differential equation without any damping term as

$$\frac{d^2 P}{dt^2} - \bar{\alpha}_1 P + \bar{\alpha}_1 P^3 - E = 0. \quad (13)$$

It should be clearly pointed out here that even though some of the tenets of the above deduction might be known in the realm of condensed matter physics, a proper theoretical basis has not been attempted so far for the derivation of a governing equation of polarization in ferroelectric materials in the context of a dynamical system from a classical Hamiltonian, by giving due importance to the Landau–Ginzburg ‘two-well’ potential. Moreover, for those engaged in the device application of such materials in the time domain analysis, the Duffing oscillator equation with and without damping could be used to explore various possibilities for deciding on the input signal.

The Klein–Gordon equation (12) can now be written with a non-dimensional damping term as

$$\frac{\partial^2 P}{\partial t^2} - \bar{k} \frac{\partial^2 P}{\partial x^2} - \bar{\alpha}_1 (P - P^3) - E + \bar{\gamma} \frac{\partial P}{\partial t} = 0 \quad (14)$$

where

$$\bar{\gamma} = \frac{\gamma P_s}{t_c E_c}. \quad (15a)$$

Here, γ is the damping coefficient, and the interaction term is defined as

$$\bar{k} = \frac{k P_s}{2 E_c}. \quad (15b)$$

For the existence of soliton solutions of the KG equation with a damping term, in order to explain the case where $\bar{\gamma} \neq 0$ and $E \neq 0$ (static electric field), a simple deduction shows the limiting or critical value of the applied electric field. Here, a situation is described to see under what condition the soliton exists in our ferroelectric system. Now, equation (14) can be rearranged as

$$\bar{k} \frac{\partial^2 P}{\partial x^2} - \frac{\partial^2 P}{\partial t^2} - \bar{\gamma} \frac{\partial P}{\partial t} + \bar{\alpha}_1 (P - P^3) = -E. \quad (16)$$

Let us take a new variable θ_1 as

$$\theta_1 = \frac{x - \sqrt{\bar{k}} \cdot V t}{\sqrt{\frac{2\bar{k}}{\bar{\alpha}_1} \cdot \sqrt{1 - V^2}}}. \quad (17)$$

By considering a component of the soliton velocity as V , let us also put

$$\frac{dP}{d\theta_1} = \phi \quad (18a)$$

$$\frac{d^2 P}{d\theta_1^2} = \frac{d\phi}{d\theta_1} = -2(P - P^3) - V_1 \phi - E_1 \quad (18b)$$

where

$$V_1 = \left(\sqrt{\frac{2}{\bar{\alpha}_1}} \right) \frac{\bar{\gamma} V}{\sqrt{1 - V^2}} \quad (19)$$

$$E_1 = \frac{2E}{\bar{\alpha}_1}. \quad (20)$$

The stationary points of a system of equations (18) are given by

$$\phi = 0 \quad (21a)$$

and

$$\bar{\alpha}_1 (P^3 - P) - E = 0. \quad (21b)$$

For the three real and distinct roots of equation (21b), the following condition has to be satisfied:

$$E < \frac{4(\bar{\alpha}_1)^2}{27} = E_{\text{crit}}(\text{say}). \quad (22)$$

Each real solution corresponds to a possible stationary state of the soliton [9]. Now, the value of E_{crit} for lithium niobate ferroelectric can be written as

$$E_{\text{crit}} = \frac{2\bar{\alpha}_1}{\sqrt{27}} = \frac{2\bar{\alpha}_1}{3\sqrt{3}} = 129.072426. \quad (23a)$$

The value of E_{crit} for lithium tantalate is

$$E_{\text{crit}} = 161.886061. \quad (23b)$$

For lithium niobate, the value for $\bar{\alpha}_1 = \frac{\alpha_1 P_s}{E_c}$ (where E_c is the coercive field) is 3.3534×10^2 . This value has been calculated by taking $\alpha_1 = 1.8849 \times 10^9 \text{ V m C}^{-1}$ from Kim *et al* [2] and $P_s = 0.75 \text{ C m}^{-2}$ and $E_c = 40 \text{ kV cm}^{-1}$ also from [2]. For lithium tantalate [2], $\bar{\alpha}_1 = 4.2058 \times 10^2$, $E_c = 17 \text{ kV cm}^{-1}$ and $P_s = 0.55 \text{ C m}^{-2}$. The values for the critical field are intentionally given to six digits after decimal point, since they show extremely high sensitivity in the dynamic system analysis [6]. These values for the electric field are the non-dimensional values, i.e. $E_{\text{crit}} = E/E_c$ (E being the dimensional value in this case), and the values of the coercive field (E_c) for both lithium niobate and lithium tantalate are known [2]. Hence, the limiting values of the (dimensional) electric field can be found as

For lithium niobate ferroelectric crystals = $516 \text{ MV m}^{-1} = 0.516 \text{ V nm}^{-1}$, and

For lithium tantalate ferroelectric crystals = $275 \text{ MV m}^{-1} = 0.275 \text{ V nm}^{-1}$.

i.e. for a ‘thin film nanodevice’ of, say, 10 nm thickness, the optimum voltages to be used are 5.16 and 2.75 V for lithium niobate and lithium tantalate crystals, respectively. The above limiting or critical values are valid for the type of coupling between the polarization domains that is assumed in the model described in this paper.

4. Conclusion

The present study of the evolution of polarization with both time and space as variables, using the Euler–Lagrange equation of motion, gives rise to the non-linear Klein–Gordon equation by taking care of coupling/interaction between the neighbouring domains of uniform polarization. When this coupling/interaction is taken as zero, the Klein–Gordon equation gives rise to a Duffing oscillator equation, when the spatial term is dropped. A non-linear KG equation can be used for dynamic analysis for soliton solutions. Eventually, a critical value of the electric field is found for lithium niobate and lithium tantalate beyond which the solitons do not exist. This limiting value of electric field may be useful for device applications in faster optical communication systems.

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