

Electrostriction and electromechanical coupling in elastic dielectrics at nanometric interfaces

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Electrostrictive coefficients, expressed in terms of dielectric parameters and elastic constants, are obtained for elastic dielectrics. In most of the recent experiments concerning determination of electrostrictive parameters in elastic dielectrics, several researchers used incorrect equations without considering the contribution from the edge effect, the shear stresses and suitable boundary conditions. This led to wrong predictions of experimental results particularly for materials with high Poisson ratios. Errors in the estimation of induced strains, varying from an underestimation of 202% to an overestimation of 168%, have been pointed out in the case of polycarbonate (PC). However, the contribution from the boundary conditions is very difficult to predict correctly and hence it is still unresolved. The electromechanical properties of the nanometric interface are also discussed. Due to significant electrostrictive and piezoelectric characteristics of the nanometric interface, PC has a great potential for electromechanical applications as electromechanical transducers, sensors and actuators.

Key words: *nanodielectric; electrostriction; elastomer*

1. Introduction

The objective of this work is to provide a clear understanding of the possible mechanism involving various dielectric and mechanical parameters for electric field induced strains in elastic dielectrics. The elastic deformation of a dielectric material under the forces exerted by electrostatic fields is called electrostriction. The mechanical deformation (stresses and strains) induced in a dielectric material (generally non piezoelectric) under the influence of an electric field occurs in two processes. The Maxwell stress effect occurs due to variation in the electric field distribution under strain, and the phenomena of electrostriction occur due to variation in the dielectric properties of the material under strain. The deformed material is no longer isotropic,

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and the scalar permittivity (ϵ) becomes a dielectric tensor (ϵ_{ij}) due to anisotropic dielectric properties, in the case of an elastic dielectric material.

In equilibrium, the total forces, i.e. the total internal body forces F_{int} and the total external forces F_{ext} such as forces due to a gravitational field, in every volume element of elastic dielectrics must be balanced to a zero value [1]:

$$F_{\text{int}} + F_{\text{ext}} = 0 \quad \text{or} \quad \sum_{j=1}^3 \frac{\partial T_{ij}}{\partial x_j} + \rho g_i = 0 \quad (1)$$

where $F_{\text{ext}} = \rho g_i$ is an external force due to a gravitational field g_i in a vertically downward direction Γ and T_{ij} is the mechanical stress tensor. If there is another external force instead of a gravitational force, then the vector ρg_i on the right hand side of above Eq. (1) must be replaced accordingly. But in the absence of external field, we have

$$\sum_{j=1}^3 \frac{\partial T_{ij}}{\partial x_j} = 0 \quad (2)$$

The strain tensor, S_{ij} , describes the state of an elastic dielectric material subjected to a small deformation, and is given as

$$S_{ij} = S_{ji} = \frac{1}{2} \left(\frac{\partial s_i}{\partial x_j} + \frac{\partial s_j}{\partial x_i} \right) \quad \text{for} \quad i \neq j \quad (3)$$

where \mathbf{s} is a displacement vector.

As the deformation is extremely small, only the first order terms in S_{ij} have been considered in the variation of components of the dielectric tensor ϵ_{ij} and the dielectric tensor is given as

$$\epsilon_{ij} = \epsilon^0 \delta_{ij} + a_1 S_{ij} + a_2 S_{kk} \delta_{ij} \quad (4)$$

where ϵ^0 is the permittivity of the undeformed body, and a_1 and a_2 are two parameters describing the variation in the dielectric properties of the material under shear and bulk deformations, respectively. The general form of the dielectric tensor, ϵ_{ij} , for the deformed material is

$$\epsilon_{ij} = \epsilon_{ij}^0 + a_{ijkl} S_{kl} \quad (5)$$

where a_{ijkl} is a constant tensor of rank four.

For linear and anisotropic dielectric under small deformation, the variation in the dielectric tensor ϵ_{ij} is given as

$$\delta\epsilon_{ij} = \sum_{k=1}^3 \sum_{l=1}^3 a_{ijkl} \delta S_{kl} \quad (6)$$

and the coefficients

$$a_{ijkl} = \frac{\partial \epsilon_{ij}}{\partial S_{kl}} \quad (7)$$

represent a tensor of rank four characteristic of dielectrics, and also have different values at different points in an inhomogeneous dielectric medium. However, all but two classes of coefficients are approximately zero, namely the body strain coefficients a_{iiii} and a_{ijij} and the shear strain coefficients a_{ijij} [1].

Electromechanical phenomena have been observed in several macroscopic systems such as polycrystalline ceramics, ceramic polymer composites, several organic polymers, biopolymers, single crystals, systems with a liquid component, etc. However, this effect is very small in the case of solid dielectrics. Experimental data on the subject are not abundant, and the researchers [2, 3] frequently used an incorrect formula in the derivation of elastic strain with respect to the Maxwell stress effect, particularly in the case of polyurethane elastomer (a cross-linked polymer), assuming various unrealistic approximations [4]. A linear electromechanical effect does not exist in the case of elastomers and Hooke's law based on thermodynamic consideration (Helmholtz free energy and Gibbs free energy concept) should not be applied for elastomers up to a large extent due to its nonlinear elastic behaviour. Electromechanical coupling effects in the case of non-piezoelectric material such as polyurethane elastomers have been exploited in the areas of fundamental sensors and actuators [5, 6]. Due to potential applications in sensing and actuation, the electrostrictive response is very important [7].

The coupling between electrical and mechanical fields at the nanometric scale has been poorly exploited up to now. The interface between two dissimilar macroscopic phases in mutual contact has invariably different properties from the bulk on the either side in continuum. The electric fields at the interface induce a mechanical stress in addition to influencing polarization and conduction phenomena. The exploitation of the interface as electromechanical transducers is the real challenge in the development of nanometric dielectrics.

2. Electrostriction in elastic dielectrics: A theoretical approach

Fundamental assumptions of the theoretical approach to electrostriction in elastic dielectrics are as follows:

- A. In unstrained state, electric and elastic properties are assumed to be isotropic.
- B. On application of an electric field, variations in permittivity are dependent on the components of strain and as a result, the material becomes electrically anisotropic.
- C. In static equilibrium, the elastic forces induced during deformations will balance the mechanical forces induced by the static field.

D. The relation between the mechanical stress (T_{ij}) and the strain (S_{ij}) is governed by Hook's law.

E. Deformation is extremely small.

F. The dependence of D on E is linear even in an anisotropic dielectric and D and E are parallel only along preferred axes, i.e.

$$D_i = \epsilon_0 \sum_{j=1}^3 \epsilon_{ij} E_j$$

where ϵ_0 is the permittivity of a free space.

The Maxwell stress tensor [8] results from a force produced by the electric field and is given as

$$T_{ij} = T_{ij}^0 + \frac{\epsilon_0}{2} (2\epsilon^0 - a_1) E_i E_j - \frac{\epsilon_0}{2} (\epsilon^0 + a_2) E^2 \delta_{ij} \quad (8)$$

where T_{ij}^0 is the stress tensor in the absence of an electric field. Generally we tend to neglect T_{ij}^0 in isotropic dielectrics but in the case of a dielectric interface, the presence of a significant number of point defects introduces distortion in the interfacial region. In the case of a size difference of two particles from two phases, elastic stress and strains are created. A larger atom introduces compressive stress and the corresponding strain around it, while a smaller interacting atom creates a tensile stress-strain field. An interstitial atom also produces strain around the void it occupies.

If the direction of the electric field is assumed to be along the X_3 axis (k direction) in the Cartesian coordinate system, the principal stresses induced electrically are

$$T_{33} = T_{33}^0 - \frac{1}{2} \epsilon_0 \epsilon^0 E^2 \left(1 - \frac{a_1 + a_2}{\epsilon^0} \right) \quad (9)$$

$$T_{11} = T_{22} = T_{11}^0 + \frac{1}{2} \epsilon_0 \epsilon^0 E^2 \left(1 + \frac{a_2}{\epsilon^0} \right) \quad (10)$$

Where $a_1 = \partial \epsilon / \partial S_1 = \partial \epsilon / \partial S_2$, due to strain in the $X_1 X_2$ plane, and $a_2 = \partial \epsilon / \partial S_3$, expresses the increment in the field direction, i.e. along the X_3 axis. T_{11}^0 and T_{33}^0 are the stresses even in the absence of an external field, however these stresses are independent of the direction in the case of isotropic dielectrics, and it should not be neglected at the interface having a significant number of point defects. However, it is very difficult to evaluate T_{ij}^0 precisely.

For a linear elastic dielectric material (described by Hooke's law), the electrically induced stresses will also generate elastic stress and strain in equilibrium condition and the elastic stress tensor T_{ij} [9] is given in terms of the elastic strain tensor, S_{ij} , Young modulus Y and Poisson's ratio, σ , by

$$T_{ij} = \frac{Y}{1+\sigma} \left(S_{ij} + \frac{\sigma}{1-2\sigma} S_{kk} \delta_{ij} \right) \quad (11)$$

conversely

$$S_{ij} = \frac{1}{Y} \left[(1+\sigma) T_{ij} - \sigma T_{kk} \delta_{ij} \right] \quad (12)$$

If the force acts only along the X_3 direction, i.e. along the direction of the electric field E , then we have $S_{11} = S_{22} = 0$, and if the sides of dielectric material are also fixed (Fig. 1), we have $S_{32} = S_{31} = 0$.

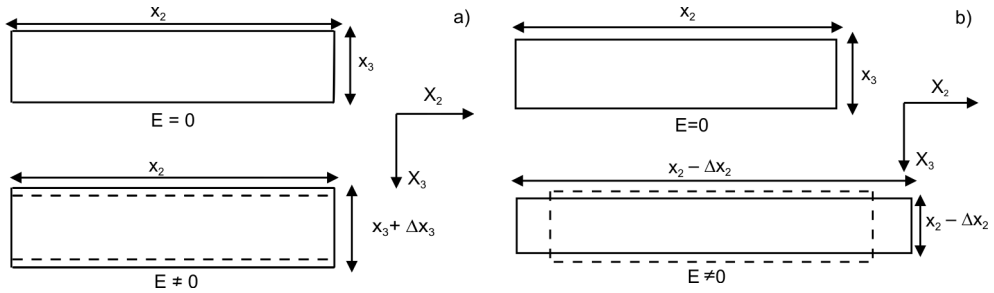


Fig. 1. Schematic diagrams illustrating the effect of electric field E (acting along the X_3 direction) on the elastic dielectric material enclosed between two parallel rigid electrodes: a) sides of a dielectric slab are rigidly fixed as a result of this; no expansion in a lateral direction takes place, i.e., $S_{31} = S_{32} = 0$, b) sides of slab are open and a longitudinal compressive strain S_{33} acts along the X_3 direction, whereas the shear strains, $S_{31} = S_{32}$, act along the X_1 and X_2 directions

For such a unilateral deformation, the principal elastic stresses and strains are

$$T_{11} = T_{22} = \frac{Y}{(1+\sigma)(1-2\sigma)} S_{33} \quad (13)$$

$$T_{33} = \frac{Y(1-\sigma)}{(1+\sigma)(1-2\sigma)} S_{33} \quad (14)$$

$$S_{33} = \frac{p(1+\sigma)(1-2\sigma)}{Y(1-\sigma)} \quad (15)$$

where $T_{33} = p$ is a compressive force, i.e. pressure. On substituting the principal value of the Maxwell stress tensor (assuming $T_{11}^0 = T_{33}^0 = 0$) from Eqs. (9) and (10) into Eq. (12), we can express the principal elastic strains as

$$S_{11} = S_{22} = \frac{1}{2} \epsilon_0 \epsilon^0 E^2 \left(1 - \frac{\sigma a_1}{\epsilon^0} + \frac{(1-2\sigma) a_2}{\epsilon^0} \right) Y^{-1} \quad (16)$$

$$S_{33} = -\frac{1}{2} \epsilon_0 \epsilon^0 E^2 \left((1+2\sigma) - \frac{a_1 + (1-2\sigma) a_2}{\epsilon^0} \right) Y^{-1} \quad (17)$$

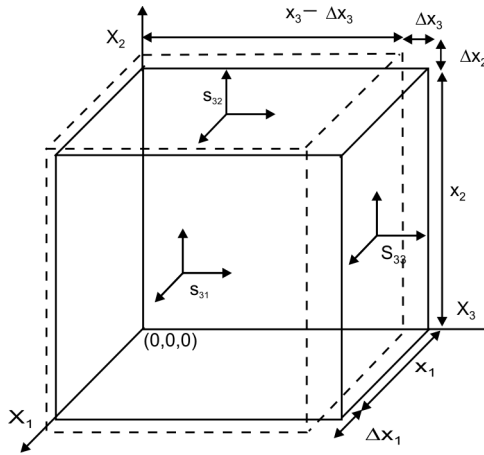


Fig. 2. Schematic diagram illustrating an elastic dielectric subjected to one normal electrical stress along the X_3 direction, causing compressive longitudinal strain S_{33} and two tensile shear strains, S_{31} and S_{32} along the X_1 and X_2 directions, respectively, as the electric field has only one component along the X_3 direction. If E also has other components, then there are other principal strains $S_{11} = S_{22} \neq 0$

The principal elastic strain, S_{33} , represents the relative change in thickness of the interface, whereas the strain, S_{11} or S_{22} , represents the relative change in the diameter of the capacitor, as described in [4], which is not correct in the case of a parallel plate capacitor in which S_{31} or S_{32} actually represent the lateral change in dimension, as E has only a component along X_3 (Fig. 2), i.e. only one normal component of stress, T_{33} , along with its two shearing components, T_{31} or T_{32} . On comparing Eqs. (16) and (17) with a general equation for electrostriction

$$S_{ij} = \gamma_{ijkl} E_k E_l \quad (18)$$

we get

$$S_{11} = S_{22} = \gamma_{31} E^2, \quad S_{33} = -\gamma_{33} E^2 \quad (19)$$

where γ_{31} and γ_{33} are the electrostrictive coefficients. A negative sign for the strain S_{33} indicates that it is a contraction along X_3 and S_{11} and S_{22} are tractions, so causing expansion along the X_1 and X_2 directions.

3. Interface as an electromechanical transducer

Macroscopic electromechanical behaviour arises from interactions occurring at the nanometric interface. At the nanodielectric interface between two phases, the segregation of charge clusters and double layer polarization are responsible for producing

electrostriction in the presence of a non-uniform electric field. If the electric field E is changed to $E + \Delta E$, the change in S_{33} becomes

$$\Delta S_{33} = -2\lambda_{33}E_0\Delta E = \mu_{33}\Delta E \quad (20)$$

where μ_{33} is the piezoelectric coefficient. Similarly, the coefficient μ_{31} and μ_{32} can be found along the X_1 and X_2 directions. The piezoelectric transverse coefficients have been frequently used [10] in piezoelectric thin film devices. The average value of compressive elastic strain induced along the X_3 direction is

$$K \int_0^\lambda \Delta S_{33} dx_3 \quad (21)$$

where λ is the Debye–Hückel length. Similarly, the expression for the tensile strains (ΔS_{32} and ΔS_{31}) along the X_1 and X_2 directions can be obtained.

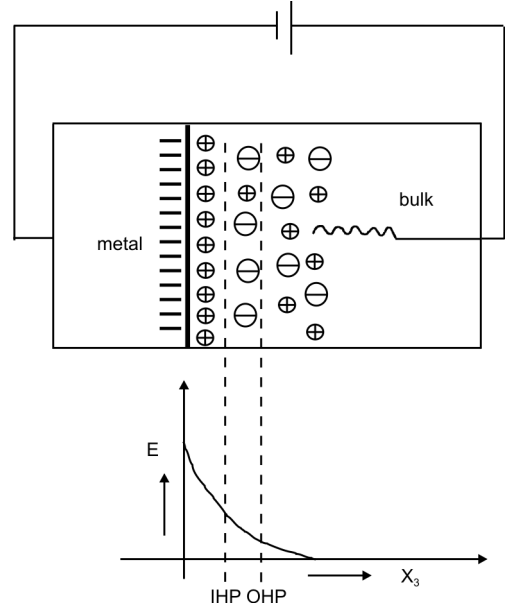


Fig. 3. Schematic diagram showing the formation of electric double layer formed due to segregation of charge clusters at an electrode–electrolyte interface. The distribution of a nonuniform electric field in noncentrosymmetric interface is also shown

At the electrode–electrolyte interface (Fig. 3) having decaying electric fields from the electrode surface, the only modification required for the calculation of E^2 is

$$\overline{E^2} = \frac{1}{\lambda} \int_0^\lambda E^2(x_3) dx_3 \quad (22)$$

The integration is over the thickness of the double layer, i.e. the Debye–Hückel length λ .

If E has only one component along the X_3 direction, we should consider the longitudinal component S_{33} and the transverse components S_{32} and S_{31} , and neglect the prin-

principal strains, S_{11} and S_{22} , as E has no components along the X_1 and X_2 direction but the actual boundary conditions satisfied (after deformations) at the interface between an electrode and dielectric are the points of main concern, and it is very difficult to correctly predict the contributions from boundary conditions. If E has small components along the X_1 and X_2 directions, we should not neglect the principal elastic strains S_{11} and S_{22} .

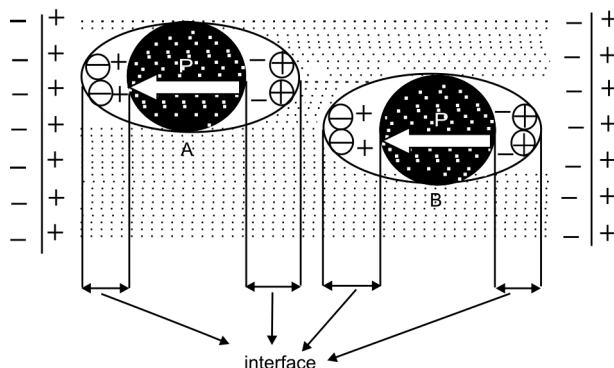


Fig. 4. Schematic diagram illustrating polycrystalline material exhibiting strong piezoelectric properties: a) for $E = 0$, dipoles are randomly oriented and double layers do not form, b) for $E \neq 0$, the material is subjected to the resulting poling process, and a number of interfaces have been formed inside the material. The interface with the double layer formation and the polarization strongly support the existence of piezoelectric characteristic

Recently piezoelectric properties have been created in a polycrystalline or composite material by using poling process. That is why an exfoliated laminar polymer (polypropylene) showed significant piezoelectric properties upon poling [11]. In micro sensor applications of PZT ceramic $\text{Pb}(\text{ZrTiO}_3)$, it has been observed that polycrystalline films with many internal interfaces exhibit a greater piezoelectric response [10, 12], whereas the epitaxial films with two electrode interfaces show a weak response. A number of nanometric structures (rings, helices, bolts etc.) in the ZnO system exhibit ordered polar non-centrosymmetry and most of them can be exploited in a wide range of piezoelectric nanometric electromechanical systems (NEMS) [13]. The above examples point to the interfacial origin of the piezoelectric effect, and these materials have many nanometric interfaces (Fig. 4) with a tremendous potential for electromechanical applications [14].

4. Discussion and comparison with previous experimental results

Errors are often committed in the derivation of suitable expressions for the interpretation of experimental results.

4.1. Contributions from shear stresses

The electromechanical response of the polyurethane elastomer (Dow 2103-80AE) has been obtained and the relative change in the thickness of a dielectric slab between the electrodes of a capacitor has been experimentally determined [3] without considering lateral stresses, the edge effect, boundary conditions and the tensor form of permittivity. The equation used may be written as:

$$\frac{\Delta x_3}{x_3} = \frac{1}{2Y} \epsilon_0 \epsilon^0 E^2 = S_{33} \quad (23)$$

However the correct mathematical expression can be seen from Eq. (17), which indicates that neglecting the lateral stress and the Poisson ratio leads to underestimation of $\Delta x_3/x_3$. Similarly, another researcher [2] used Maxwell's principal stress (Eq. (9)) for the determination of relative change in the thickness of a dielectric slab, neglecting the contributions from lateral elastic stresses and the edge effect. Here, the expression used was

$$\frac{\Delta x_3}{x_3} = -\frac{1}{2Y} \epsilon_0 \epsilon^0 E^2 \left(1 - \frac{a_1 + a_2}{\epsilon^0} \right) \quad (24)$$

Equation (17) gives the correct representation of the expression for elastic dielectrics. In particular, the elastomers, which undergo recoverable deformations of a few hundred percent, do not exhibit linear elastic behaviour (Fig. 5) and do not follow Hooke's law, in contrast to an ordinary elastic material. Thus the experimental results [2, 3] for the cross linking elastomers cannot be correctly predicted from the actual induced strain response using Eqs. (23) and (24).

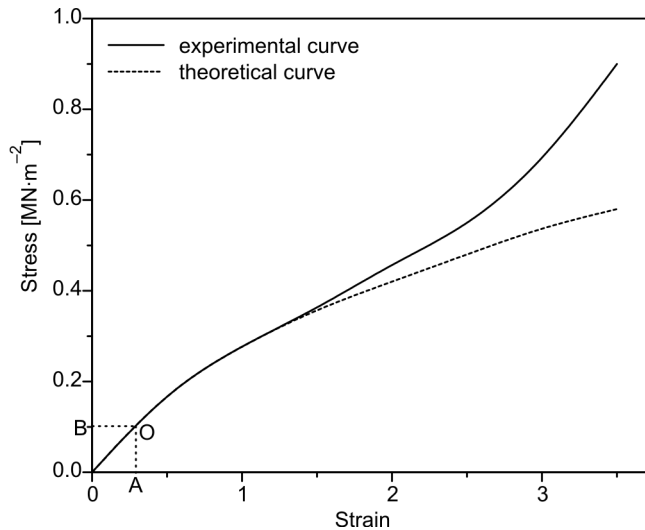


Fig. 5. Nonlinear stress-strain characteristic which does not follow Hooke's law, however, Hooke's law is considered for very small deformation up to the point O

Theoretically, Poisson's ratio (σ) varies from -1 to 0.5 but in practice no material is available with $\sigma < 0$ [9]. Poisson's ratio for polymers has been found to be ca. 0.5 [15]. Neglect of lateral stresses, particularly in the case of elastic dielectrics or polymers with high Poisson's ratio, and using Eq. (9) leads to incorrect estimation of the electrically induced strain. On putting $\sigma = 0.5$ in Eq. (17), we get

$$S_{33} = -\frac{1}{2}\epsilon_0\epsilon^0 E^2 \left[2 - \frac{a_1}{\epsilon_0} \right] Y^{-1} \quad (25)$$

and this is quite different from Eq. (9), in the sense that the contribution from the term a_2 has been cancelled due to the presence of the $(1 - 2\sigma)$ term in Eq. (17). Actually the longitudinal principal strain S_{33} calculated from Eq. (25) is twice the value of the strain calculated in [3] from Eq. (23) for a material with $\sigma = 0.5$ and $a_1 = 0$. Errors in the estimations of induced strains can be pointed out in the case of polycarbonate (PC). The following data for polycarbonate are given [16]: $|a_1| = 1.404$, $|a_2| = 2.6$, $|a_2| = 2.6$, $\epsilon^0 = 2.9^*$ at 1 MHz, $\sigma = 0.37$ at 1 MHz.

Comparing Eqs. (23) and (24) with Eq. (17), after substituting the above data, an overestimation of 168% and an underestimation of 202% , respectively, have been found.

The use of the correct Eq. (17) is necessary, particularly for the material having a high Poisson's ratio. Using the dielectric constant and the elastic compliance data, the contribution of the Maxwell stress to the total strain response can be determined.

If the sides of the dielectric slab/interface are rigidly fixed, the effective stiffness (T_{33}/S_{33}) is governed by Eq. (14). The dielectric parameters, a_1 and a_2 , are experimentally obtained [16] by determining $d\epsilon_{33}/dS_{33}$ in constrained and unconstrained conditions, and using Eq. (4). However, the linearity of the theory of electrostriction fails if the deformations are not quadratic in the applied field, particularly at a higher field strength.

In the case of dielectrics with a non-uniform field, e.g. cylindrical capacitor and nanometric interface, we should use either Eq. (22) or deal with the problem separately to evaluate $\overline{E^2}$ for the situation. In the presence of a non-uniform electric field with a high field strength, the phenomenon of dielectrophoresis takes place. However, it is in general a weak effect, particularly in the case of a solid dielectric.

4.2. Contributions from edge effects

Due to free edges of plates of a capacitor, the distribution of charge over them is not uniform. Hence a correction due to edge effects [8] is very much required for all practical purposes in which the thickness of the dielectric slab is comparable to its

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lateral dimension i.e., $\Delta x_3 \approx (\text{cross sectional area of plate})^{1/2}$. However, this correction may be neglected in the case of a thin film capacitor and a nanometric interface having a lateral dimension that is much greater relative to its thickness (typically of order a few nm).

4.3. Contribution from boundary conditions

For experimental study [3, 16], a parallel plate capacitor with a dielectric film/slab has been considered, due to its simple, symmetrical geometry. However, due to electrostrictive deformation, the permittivity of different regions (Fig. 1 shows two regions, one with an air gap and other with a dielectric medium) is different, thus the solution to the field or potential must be different although having the same general form of solution (to the Laplace equation). Appropriate electrical boundary conditions must be satisfied at the interface between the two regions.

For the mechanical boundary conditions, Eq. (1) must be satisfied in equilibrium, i.e. the external forces on the boundary may be regarded as a continuation of the internal stress distribution.

However, it is very difficult to predict exact boundary conditions at the interface between electrodes and the dielectric or between two regions with different permittivities. If the field is applied exactly along the X_3 direction, the strain components, S_{33} (normal/bulk strain along X_3), and S_{31} and S_{32} (shear strains along the X_1 and X_2 directions) play the major role in the electromechanical phenomena exhibited by the interface in nanometric dielectrics [17].

5. Conclusion

In elastic dielectrics, shear and lateral stresses play a major role in the correct estimation of induced elastic strain. Thus neglecting the contribution from lateral and shear stresses leads to overestimation or underestimation of the results. However, the boundary conditions at the interface between electrode and the dielectrics are the points of main concern, as it is very difficult to predict exact mechanical and electrical boundary conditions in deformed dielectrics. It has also been pointed out that nanometric dielectric interfaces, which are non-centrosymmetric systems, have tremendous potential for electromechanical applications. A lot of work has to be done, particularly in the field of biology, cellular polymers, biopolymers, intercellular biomembranes, etc. at the nanoscale level, in order to exploit nanometric interfaces for various applications as sensors, actuators and transducers.

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