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Electric Field Gradient Theory with Surface Effect for Nano-Dielectrics

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Abstract: The electric field gradient effect is very strong for nanoscale dielectrics. In addition, neither the surface effect nor electrostatic force can be ignored. In this paper, the electric Gibbs free energy variational principle for nanosized dielectrics is established with the strain/electric field gradient effects, as well as the effects of surface and electrostatic force. As regards the surface effects both the surface stress and surface polarization are considered. From this variational principle, the governing equations and the generalized electromechanical Young-Laplace equations, which take into account the effects of strain/electric field gradient, surface and electrostatic force, are derived. The generalized bulk and surface electrostatic stress are obtained from the variational principle naturally. The form are different from those derived from the flexoelectric theory. Based on the present theory, the size-dependent electromechanical phenomenon in nano-dielectrics can be predicted.

Keywords: Electric field gradient; Variational principle; Strain gradient; Surface polarization; Electrostatic stress; Surface effect

1 Introduction

The development of nanotechnology brings on thinner and thinner dielectric films and other nanosized electronic devices. With miniaturization of a dielectric material down to nanoscale, the size effects become dominant. Although piezoelectricity has attracted many researchers (Chen *at al.*, 2009; Apte, Ganguli, 2009; Ma, Wu, 2009; Wu, Huang, 2009; Wu *et al.*, 2008; Wu, Liu, 2007; Wu *et al.*, 2005; Cheng, Chen, 2004), the classical electromechanical coupling theory fails to describe the size-dependent phenomenon. It is well known that both surface theory and gradient theory can describe the size effects. However, the theory concerned

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with both gradient and surface effects is still lacked, especially for the electromechanical coupling problems where the gradient effects of electric variables often play an important role. Shen and Hu (2010), and Hu and Shen (2010) developed a theory for nanosized dielectrics with the flexoelectric effects as well as the surface effects. For dielectrics, the polarization gradient theory (Mindlin, 1969) and the electric field gradient theory (Landau and Lifshitz, 1984) can be considered as theories for weak nonlocal effects (Maugin, 1979; Yang, 2006). The electric field gradient theory is also called dielectrics with spatial dispersion, and is equivalent to the theory of dielectrics with electric quadrupoles (Kafadar, 1971) due to that the electric quadrupole is the thermodynamic conjugate of the electric field gradient. Theories for elastic dielectrics with electric quadrupoles are also discussed in Demiray & Eringen (1973), Maugin (1979, 1980), and Eringen & Maugin (1990). Kalpakides and Agiasofitou (2002) developed a formulation including both strain gradient and electric field gradient. The duality between the theory of electric field-gradient and flexoelectric theory can be found in Maugin (1980). Yang *et al.* (2004), and Yang & Yang (2004) showed that due to the electric field gradient the capacitance of a thin film deviates from the classical value when the film thickness approaches a microscopic characteristic length, and that plane waves become dispersive when the wave length approaches a microscopic characteristic length. Yang (2004b) examined the effect of electric field gradient on a semi-infinite crack in polarized ceramics. Yang *et al.* (2005) studied the field concentration near a small hole and found that the concentration depends on the size of the hole because of the electric field gradient. Yang *et al.* (2006) analyzed the effects of electric field gradient on the anti-plane problem of a small, circular inclusion in polarized ceramics. These papers demonstrated that the electric field gradient exhibits a size effect and has significant impact on the apparent piezoelectric and elastic behavior.

However, the surface effect and electrostatic force are not taken into consideration in these papers. For dielectrics, the surface effect includes both surface stress and surface polarization which is induced due to the dangling bonds on the surface. The rearrangement of the bonding at surfaces causes a charge displacement that may greatly alter the polarization properties of the surface with respect to the bulk (Brandino and Cicero, 2007). For nanosized dielectrics, the ratio of surface area to volume becomes high, so the surface polarization and stress turn out to be significant. In Slavchov *et al.* (2006), the effect of the surface polarization was studied with the help of a definition of surface dielectric constants for surface. The effect of the surface polarization in polar perovskites was investigated by means of the first principles in Fechner *et al.* (2008). Camacho and Nossa (2009) showed that the influences of surface polarization on the dielectric properties of quantum dots arrays are significant. Consequently, it is necessary to develop a theoritical framework to concern with both surface stress and surface polarization for nanosized dielectrics. In NEMS design, the electrostatic force also plays a very important role. Many papers (Gao *et al*., 2001; Dequesnes *et al*., 2002, 2004; Lee, Chen, 2009) demonstrated that the effects of the electrostatic forces are very strong in nanoscale even for linear elastic problems. Hence, appropriate consideration of the electrostatic force on the nanostructures is very important in the analysis of nanoscale electromechanical coupling problems.

The variational principles have been regarded as the bases of the analysis and computations for electromechanical problems in dielectric materials for a long time. Toupin (1956) gave a variational principle for a linear piezoelectric material; Kuang (2007, 2008a, b, 2009) systematically discussed the thermodynamic nonlinear variational principles with electric Gibbs free energy and internal energy for nonlinear problems (Shen *et al*., 2000). From these variational principles the complete governing equation systems were deduced and the Maxwell stress were naturally derived. However, all these papers did not consider the strain/electric field gradient and surface effects.

In this paper, the variational principle for nanosized dielectrics with strain/electric field gradient and surface effects are established. Based on the variational principle, a theoretical framework is formulated to examine the size effect due to both electric field gradient and surface effects for a dielectric material. The surface effect includes the effect of surface stress and surface polarization. The governing equations, which include the effect of strain/electric field gradient and the electrostatic force, and the generalized electromechanical Young-Laplace equations for a electric field gradient dielectric material with surface effect are presented. The generalized bulk and surface electrostatic stress can be obtained from the variational principle naturally.

2 The bulk and surface electric Gibbs free energy

In Gurtin and Murdoch (1975), the influence of surface effect on stress and strain fields is formulated in a continuum framework for elastic surface of solid. The surface constitutive relations together with the surface conditions of the stress provide the necessary conditions for the boundary-value problem to determine the stress and strain fields with surface effect. In this paper, we will develop the continuum framework for nano-dielectrics to formulate the influence of surface and strain/electric field gradient effects on electromechanical coupling field. In the gradient dielectric material model, a surface element at a material point can transmit not only stress and higher order stress (moment stress) but also the electric displacement and electric quadrupole. So in the electric field gradient electromechanical theory, three traditional displacements, and an electric potential are used to describe the deformable point particles. For the electric field gradient electromechanical theory with surface effect, additional surface electromechanical constitutive relations and the surface conditions are needed. In order to simplify the discussion in this paper, the infinitesimal deformation is assumed.

To take the surface effects into account, the electric Gibbs free energy can be decomposed into two parts, *i.e.*, the surface and bulk parts. Within the assumption of an extended linear theory for dielectrics incorporating terms involving the gradients of the strain and the electric field, the most general expression for the bulk electric Gibbs free energy density function U_b can be written as

$$\begin{aligned} U_b =& -\frac{1}{2}a_{kl}E_kE_l - \frac{1}{2}b_{ijkl}E_{i,j}E_{k,l} + \frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} - e_{ijkl}\varepsilon_{ij}E_{k,l} - d_{ijk}\varepsilon_{ij}E_k - h_{ijk}E_iE_{j,k} \\ & - f_{ijkl}E_iu_{j,kl} + r_{ijklm}\varepsilon_{ij}u_{k,lm} - \eta_{ijkmn}E_{i,j}u_{k,mn} + \frac{1}{2}g_{ijklmn}u_{i,jk}u_{l,mn} \\ =& -\frac{1}{2}a_{kl}E_kE_l - \frac{1}{2}b_{ijkl}V_{ij}V_{kl} + \frac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} - e_{ijkl}\varepsilon_{ij}V_{kl} - d_{ijk}\varepsilon_{ij}E_k - h_{ijk}E_iV_{jk} \\ & - f_{ijkl}E_iw_{jkl} + r_{ijklm}\varepsilon_{ij}w_{klm} - \eta_{ijkmn}V_{ij}w_{kmn} + \frac{1}{2}g_{ijklmn}w_{ijk}w_{lmn} \end{aligned} \tag{1}$$

where $\mathbf{a}$, $\mathbf{b}$, $\mathbf{c}$, $\mathbf{e}$, $\mathbf{d}$, $\mathbf{h}$, $\mathbf{f}$, $\mathbf{r}$, $\boldsymbol{\eta}$, and $\mathbf{g}$ are the material property tensors. Particularly, $\mathbf{a}$ and $\mathbf{c}$, are the second-order permittivity and four-order elastic constant tensors, respectively. The tensor $\mathbf{g}$ represents the purely nonlocal elastic effects and corresponds to the strain gradient elasticity theories. $\mathbf{u}$ is the displacement, the comma indicates differentiation with respect to the spatial variables. $\boldsymbol{\varepsilon}$ is the strain tensor and $\mathbf{E}$ is the electric field tensor, which are defined, respectively, as

$$\varepsilon_{ij} = \frac{1}{2}\left(u_{i,j} + u_{j,i}\right), \tag{2a}$$

i.e.,

$$\boldsymbol{\varepsilon} = \frac{1}{2}\left(\nabla\otimes\mathbf{u} + \mathbf{u}\otimes\nabla\right) \tag{2b}$$

$$E_i = -\varphi_{,i},$$

i.e.,

$$\mathbf{E} = -\nabla\varphi$$

where φ is the electrostatic potential. $\mathbf{w}$ and $\mathbf{V}$ are the strain gradient tensor and the electric field gradient tensor respectively, which are defined as

$$w_{ijm} = u_{i,jm}, \quad \mathbf{w} = \frac{1}{2}\nabla\otimes\left(\nabla\otimes\mathbf{u} + \mathbf{u}\otimes\nabla\right) \tag{3}$$

$$V_{ij} = E_{i,j}, \quad \mathbf{V} = -\nabla \otimes \nabla \varphi \tag{4}$$

Thus, we have $\varepsilon_{ij} = \varepsilon_{ji}$, $w_{ijm} = w_{jim}$, and $V_{ij} = V_{ji}$.

The material property tensors **a**, **b**, **c**, **e**, **d**, **h**, **f**, **r**, $\boldsymbol{\eta}$, and **g** may be taken different value for different material. If the electromechanical coupling is not taken into account, all the material property tensors responding to the electromechanical coupling are equal to 0, *i.e.*, **e**, **d**, **f**, and $\boldsymbol{\eta}$ are 0. For the centrosymmetric dielectric, **d** and $\boldsymbol{\eta}$ are equal to 0. If the strain gradient is not considered (only the electric field gradient is considered), **f**, **r**, $\boldsymbol{\eta}$, and **g** are all zero, which is reduced to that in Yang (2004b).

Under the infinitesimal deformation, the constitutive equations for the bulk can be expressed in terms of the electric Gibbs free energy as

$$\sigma_{ij} = \frac{\partial U_b}{\partial \varepsilon_{ij}} = c_{ijkl}\varepsilon_{kl} - e_{ijkl}V_{kl} - d_{ijk}E_k + r_{ijklm}w_{klm} \tag{5a}$$

$$\tau_{ijm} = \frac{\partial U_b}{\partial w_{ijm}} = -f_{kijm}E_k + r_{klijm}\varepsilon_{kl} - \eta_{klijm}V_{kl} + g_{ijmknl}w_{knl} \tag{5b}$$

$$D_i = -\frac{\partial U_b}{\partial E_i} = a_{ij}E_j + d_{jki}\varepsilon_{jk} + h_{ijk}V_{jk} + f_{ijkl}w_{jkl} \tag{5c}$$

$$Q_{ij} = -\frac{\partial U_b}{\partial V_{ij}} = b_{ijkl}V_{kl} + e_{klij}\varepsilon_{kl} + h_{kij}E_k + \eta_{ijkmn}w_{kmn} \tag{5d}$$

where σ_{ij} is the stress tensor which is same as that in classical elasticity, D_i is the electric displacement vector, and τ_{ijm} and Q_{ij} are the higher order stress (moment stress) and electric quadrupole, respectively. It is noted that $\sigma_{ij} = \sigma_{ji}$, $\tau_{ijm} = \tau_{jim}$ and $Q_{ij} = Q_{ji}$.

By means of equations (5a-d), the bulk electric Gibbs free energy density function U_b can be rewritten as

$$\begin{aligned} U_b =& \frac{1}{2}\sigma_{ij}\varepsilon_{ij} + \frac{1}{2}\tau_{ijm}u_{i,jm} - \frac{1}{2}D_iE_i - \frac{1}{2}Q_{ij}V_{ij} \\ =& \frac{1}{2}\boldsymbol{\sigma}:\boldsymbol{\varepsilon} + \frac{1}{2}\boldsymbol{\tau}\,\vdots\,\mathbf{w} - \frac{1}{2}\mathbf{D}\cdot\mathbf{E} - \frac{1}{2}\mathbf{Q}:\mathbf{V} \end{aligned} \tag{6}$$

The residual surface stress affects the elastic properties of nanostructure significantly. Hence, in this paper, the residual surface tension and residual surface charge are considered. The surface internal energy density $U_s\left(\boldsymbol{\varepsilon}_s, \mathbf{w}_s, \mathbf{E}_s, \mathbf{V}_s\right)$ is a smooth nonlinear function of the surface strain, surface electric field, and their first gradients, which can be expressed into a series expansion

$$
\begin{aligned}
U_s =& U_{s0} - \omega_\alpha E^s_\alpha - \kappa_{\alpha\beta} V^s_{\alpha\beta} + \Gamma_{\alpha\beta}\varepsilon^s_{\alpha\beta} + \Omega_{\alpha\beta\gamma} w^s_{\alpha\beta\gamma} - \frac{1}{2} a^s_{\alpha\beta} E^s_\alpha E^s_\beta \\
& - \frac{1}{2} b^s_{\alpha\beta\gamma\kappa} V^s_{\alpha\beta} V^s_{\gamma\kappa} + \frac{1}{2} c^s_{\alpha\beta\gamma\kappa}\varepsilon^s_{\alpha\beta}\varepsilon^s_{\gamma\kappa} - e^s_{\alpha\beta\gamma\kappa}\varepsilon^s_{\alpha\beta} V^s_{\gamma\kappa} - d^s_{\alpha\beta\gamma}\varepsilon^s_{\alpha\beta} E^s_\gamma - h^s_{\alpha\beta\gamma} E^s_\alpha V^s_{\beta\gamma} \\
& - f^s_{\alpha\beta\gamma\kappa} E^s_\alpha w^s_{\beta\gamma\kappa} + r^s_{\alpha\beta\gamma\kappa\lambda}\varepsilon^s_{\alpha\beta} w^s_{\gamma\kappa\lambda} - \eta^s_{\alpha\beta\gamma\kappa\lambda} V^s_{\alpha\beta} w^s_{\gamma\kappa\lambda} + \frac{1}{2} g^s_{\alpha\beta\gamma\kappa\lambda\tau} w^s_{\alpha\beta\gamma} w^s_{\kappa\lambda\tau}
\end{aligned}
\tag{7}
$$

where U_{s0}, ω_α, $\kappa_{\alpha\beta}$, $\Gamma_{\alpha\beta}$, $\Omega_{\alpha\beta\gamma}$, $a^s_{\alpha\beta}\ldots$ are material constants depended on surface, which can be determined by either experiments or atomistic simulations. The property of all these material constants can be determined according to Aris (1962), for example, $\Gamma_{\alpha\beta}$ is isotropic if and only if $\Gamma_{\alpha\beta} = \Gamma_{11}\delta_{\alpha\beta}$. ω_α, $\kappa_{\alpha\beta}$, $\Gamma_{\alpha\beta}$, and $\Omega_{\alpha\beta\gamma}$ give the residual surface electric displacement, residual surface electric quadrupole, residual surface stress, and residual surface higher order stress, respectively. In this paper, both the superscript and subscript 's' represent the quantity on the surface, and the Greek indices run from 1 to 2 while the Latin indices run from 1 to 3. Here, we only give the low-order terms. In the case of infinitesimal deformation, the high-order terms (>2) can be ignored. Then the linear surface constitutive relation can be expressed as

$$\boldsymbol{\sigma}_s = \frac{\partial U_s}{\partial \boldsymbol{\varepsilon}_s}, \quad \boldsymbol{\tau}_s = \frac{\partial U_s}{\partial \mathbf{w}_s} \tag{8a}$$

$$\mathbf{D}_s = -\frac{\partial U_s}{\partial \mathbf{E}_s}, \quad \mathbf{Q}_s = -\frac{\partial U_s}{\partial \mathbf{V}_s} \tag{8b}$$

or

$$\sigma^s_{\alpha\beta} = \frac{\partial U_s}{\partial \varepsilon^s_{\alpha\beta}} = \Gamma_{\alpha\beta} + c^s_{\alpha\beta\gamma\kappa}\varepsilon^s_{\gamma\kappa} - e^s_{\alpha\beta\gamma\kappa} V^s_{\gamma\kappa} - d^s_{\alpha\beta\gamma} E^s_\gamma + r^s_{\alpha\beta\gamma\kappa\lambda} w^s_{\gamma\kappa\lambda} \tag{9a}$$

$$\tau^s_{\alpha\beta\gamma} = \frac{\partial U_s}{\partial w^s_{\alpha\beta\gamma}} = \Omega_{\alpha\beta\gamma} - f^s_{\alpha\beta\gamma\kappa} E^s_\kappa + r^s_{\alpha\beta\gamma\kappa\lambda}\varepsilon^s_{\kappa\lambda} - \eta^s_{\kappa\lambda\alpha\beta\gamma} V^s_{\kappa\lambda} + g^s_{\alpha\beta\gamma\kappa\lambda\tau} w^s_{\kappa\lambda\tau} \tag{9b}$$

$$D^s_\alpha = -\frac{\partial U_s}{\partial E^s_\alpha} = \omega_\alpha + a^s_{\alpha\beta} E^s_\beta + d^s_{\beta\gamma\alpha}\varepsilon^s_{\beta\gamma} + h^s_{\alpha\beta\gamma} V^s_{\beta\gamma} + f^s_{\alpha\beta\gamma\kappa} w^s_{\beta\gamma\kappa} \tag{9c}$$

$$Q^s_{\alpha\beta} = -\frac{\partial U_s}{\partial V^s_{\alpha\beta}} = \kappa_{\alpha\beta} + b^s_{\alpha\beta\gamma\kappa} V^s_{\gamma\kappa} + e^s_{\gamma\kappa\alpha\beta}\varepsilon^s_{\gamma\kappa} + h^s_{\gamma\alpha\beta} E^s_\gamma + \eta^s_{\alpha\beta\gamma\kappa\lambda} w^s_{\gamma\kappa\lambda} \tag{9d}$$

where $\boldsymbol{\sigma}_s$ and $\boldsymbol{\tau}_s$ are the surface stress and surface higher order stress tensors, $\mathbf{D}_s$ and $\mathbf{Q}_s$ are the surface electric displacement and surface electric quadrupole, respectively. $\boldsymbol{\varepsilon}_s$ and $\mathbf{w}_s$ are the surface strain and surface strain gradient, $\mathbf{E}_s$ and $\mathbf{V}_s$ are the surface electric field and the surface electric field gradient, respectively. The surface electric displacement $\mathbf{D}_s$ comes from the surface polarization which is due to the dangling bonds on the surface. The rearrangement of the bonding at surfaces leads to a charge displacement that may deeply modify the polarization properties of the surface with respect to the bulk (Brandino and Cicero, 2007). The unit of surface electric displacement is C/m, which means the dipole moment per unit area and was used in Brandino and Cicero (2007), while that of the bulk electric displacement is C/m^2. In the surface electromechanical model, the electrostatic potential φ is continuous across the surface same as the displacement, so the electric field $\mathbf{E}_s$ of the surface can be defined. By means of equations (9a-d), the surface internal energy density function U_s can be rewritten as

$$\begin{aligned} U_s =& U_{s0} + \frac{1}{2}\Gamma_{\alpha\beta}\varepsilon^s_{\alpha\beta} + \frac{1}{2}\Omega_{\alpha\beta\gamma}w^s_{\alpha\beta\gamma} - \frac{1}{2}\omega_\alpha E^s_\alpha - \frac{1}{2}\kappa_{\alpha\beta}V^s_{\alpha\beta} \\ &+ \frac{1}{2}\sigma^s_{\alpha\beta}\varepsilon^s_{\alpha\beta} + \frac{1}{2}\tau^s_{\alpha\beta\gamma}w^s_{\alpha\beta\gamma} - \frac{1}{2}D^s_\alpha E^s_\alpha - \frac{1}{2}Q^s_{\alpha\beta}V^s_{\alpha\beta} \\ =& U_{s0} + \frac{1}{2}\boldsymbol{\Gamma} : \boldsymbol{\varepsilon}_s + \frac{1}{2}\boldsymbol{\omega} \vdots \mathbf{w}_s - \frac{1}{2}\boldsymbol{\omega}\cdot\mathbf{E}_s - \frac{1}{2}\boldsymbol{\kappa} : \mathbf{V}_s \\ &+ \frac{1}{2}\boldsymbol{\sigma}_s : \boldsymbol{\varepsilon}_s + \frac{1}{2}\boldsymbol{\tau}_s \vdots \mathbf{w}_s - \frac{1}{2}\mathbf{D}_s\cdot\mathbf{E}_s - \frac{1}{2}\mathbf{Q}_s : \mathbf{V}_s \end{aligned} \tag{10}$$

A curvilinear coordinate system with covariant base vector $\boldsymbol{\xi}_\alpha$ ($\alpha = 1,2$) on the tangent plane of the surface is constructed on the surface,. The unit normal vector is denoted by $\boldsymbol{\xi}_3$ or $\mathbf{n}$. The surface strain $\boldsymbol{\varepsilon}_s$ is a second rank tensor in a two-dimensional space, and can be considered as the projection of the tensor $\boldsymbol{\varepsilon}$ in the three-dimensional space onto the tangent plane. The strain tensor in a three-dimensional space can be expressed as

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_s + \varepsilon_{\alpha 3}\boldsymbol{\xi}_\alpha\otimes\boldsymbol{\xi}_3 + \varepsilon_{3\beta}\boldsymbol{\xi}_3\otimes\boldsymbol{\xi}_\beta + \varepsilon_{33}\boldsymbol{\xi}_3\otimes\boldsymbol{\xi}_3, \quad \alpha,\ \beta = 1,2 \tag{11}$$

where $\boldsymbol{\varepsilon}_s = \varepsilon_{\alpha\beta}\boldsymbol{\xi}_\alpha\otimes\boldsymbol{\xi}_\beta$. The same relationships exist between other surface tensors and their corresponding bulk tensors. Analogous to Chen et al. (2007), the surface strain $\boldsymbol{\varepsilon}_s$ and surface strain gradient $\mathbf{w}_s$ can be expressed as

$$\boldsymbol{\varepsilon}_s = \frac{1}{2}\left(\nabla_s\otimes\mathbf{u} + \mathbf{u}\otimes\nabla_s\right), \quad \mathbf{w}_s = \frac{1}{2}\nabla_s\otimes\left(\nabla_s\otimes\mathbf{u} + \mathbf{u}\otimes\nabla_s\right) \tag{12}$$

where ∇_s denotes the gradient operator of the surface, and the surface electric field $\mathbf{E}_s$ and the surface electric field gradient $\mathbf{V}_s$ can be expressed as

$$\mathbf{E}_s = -\nabla_s\varphi, \quad \mathbf{V}_s = -\nabla_s\otimes\nabla_s\varphi \tag{13}$$

The displacement $\mathbf{u}$ on the surface can be decomposed into a tangential part $\mathbf{u}_t$ and a normal part $\mathbf{u}_n$ as

$$\mathbf{u} = \mathbf{u}_t + \mathbf{u}_n \tag{14}$$

where

$$\mathbf{u}_t = \mathbf{R} \cdot \mathbf{u}_s, \quad \mathbf{u}_n = u^n \mathbf{n} \tag{15}$$

with the projector tensor $\mathbf{R} = \mathbf{I} - \mathbf{n} \otimes \mathbf{n}$, $\mathbf{I}$ is the unit tensor and $\mathbf{n}$ is the unit normal vector. By using the Weingarten formula (Gurtin and Murdoch, 1975), it can be obtained that

$$\nabla_s \otimes \mathbf{u}_n = -u^n \mathbf{k} \tag{16}$$

where $\mathbf{k}$ is the curvature tensor of the surface. Then the surface strain $\boldsymbol{\varepsilon}_s$ and surface strain gradient $\mathbf{w}_s$ can be expressed respectively as

$$\boldsymbol{\varepsilon}_s = \nabla_s \otimes \mathbf{u}_t - u^n \mathbf{k} \tag{17}$$

$$\mathbf{w}_s = \nabla_s \otimes \nabla_s \otimes \mathbf{u}_t - \nabla_s \otimes (u^n \mathbf{k}) \tag{18}$$

3 The governing equations and generalized electromechanical Young-Laplace equations

Now, we consider a dielectric material with surface and gradient (strain/electric field gradient) effects, which is subjected to a displacement and electric potential boundary and the body force and body charge density are neglected. Assuming that the dielectric material occupies a volume V bounded by a surface a, then, analogous to that in Eringen & Maugin (1990), the variational principles can be taken as

$$\delta \int_V U_b dV + \delta \int_a U_s da = 0 \tag{19}$$

where δ is the variational sign.

According to the Reynold's transport theorem (Kuang, 2002; Ogden, 1984), we have

$$\delta \int_V U_b dV = \int_V \delta U_b dV + \int_V U_b \delta (\nabla \cdot \mathbf{u}) dV \tag{20}$$

$$\delta \int_a U_s da = \int_a \delta U_s da + \int_a U_s \delta (\nabla_s \cdot \mathbf{u}) da \tag{21}$$

At first, we consider the variation of the bulk electric Gibbs free energy, which is followed that

$$\begin{aligned}\delta\int_V U_b dV &= \int_V \delta U_b dV + \int_V U_b \delta u_{k,k} dV \\ &= \int_V \left(\sigma_{ij}\delta u_{i,j} + \tau_{ijm}\delta u_{i,jm} + D_i \delta\varphi_{,i} + Q_{ij}\delta\varphi_{,ij}\right) dV \\ &\quad + \int_V \frac{1}{2}\left(\sigma_{ij}u_{i,j} + \tau_{ijm}u_{i,jm} + D_i\varphi_{,i} + Q_{ij}\varphi_{,ij}\right)\delta u_{k,k} dV\end{aligned} \tag{22}$$

As pointed out in Kuang (2007, 2008a, b, 2009), the virtual displacement not only causes the variation of strain, but also causes the variation of electric potential and its gradients, then we can obtain that

$$\delta\varphi = \delta_\varphi\varphi + \delta_u\varphi = \delta_\varphi\varphi + \varphi_{,j}\delta u_j \tag{23}$$

$$\delta\varphi_{,i} = \delta_\varphi\varphi_{,i} + \delta_u\varphi_{,i} = \delta_\varphi\varphi_{,i} + \varphi_{,ij}\delta u_j = \delta_\varphi\varphi_{,i} + \varphi_{,ji}\delta u_j \tag{24}$$

$$\delta\varphi_{,ij} = \delta_\varphi\varphi_{,ij} + \delta_u\varphi_{,ij} = \delta_\varphi\varphi_{,ij} + \varphi_{,ijk}\delta u_k = \delta_\varphi\varphi_{,ij} + \varphi_{,ikj}\delta u_k \tag{25}$$

where $\delta_\varphi\varphi$, $\delta_\varphi\varphi_{,i}$, and $\delta_\varphi\varphi_{,ij}$ are produced by the virtual electric potential, $\delta_u\varphi = \varphi_{,j}\delta u_j$, $\delta_u\varphi_{,i} = \varphi_{,ij}\delta u_j = \varphi_{,ji}\delta u_j$ and $\delta_u\varphi_{,ij} = \varphi_{,ijk}\delta u_k = \varphi_{,ikj}\delta u_k$ are produced by the virtual displacement. Equations (23)-(25) is somewhat analogous to the Landau and Lifshitz's theory (1984) and Stratton (1941), where they pointed out that the virtual displacements not only produce the variation, but also produce the variation of electric potential.

Using the Gauss divergence theorem and relations (23)-(25), the integrals in the first part of equation (22) can be reduced to as following

$$\begin{aligned}\int_V \left(\sigma_{ij}\delta u_{i,j} + \tau_{ijm}\delta u_{i,jm}\right) dV &= \int_a \sigma_{ij}n_j\delta u_i da - \int_V \sigma_{ij,j}\delta u_i dV \\ &\quad + \int_a \tau_{ijm}n_m\delta u_{i,j} da - \int_a \tau_{ijm,m}n_j\delta u_i da + \int_V \tau_{ijm,mj}\delta u_i dV\end{aligned} \tag{26}$$

$$
\begin{aligned}
\int_V D_i \delta \varphi_{,i} dV &= \int_V \left(D_i \delta_\varphi \varphi_{,i} + D_i \varphi_{,ji} \delta u_j \right) dV \\
&= \int_a n_i D_i \delta_\varphi \varphi da - \int_V D_{i,i} \delta_\varphi \varphi dV + \int_a n_i D_i \varphi_{,j} \delta u_j da - \int_V \varphi_{,j} \left(D_i \delta u_j \right)_{,i} dV \\
&= \int_a n_i D_i \delta_\varphi \varphi da - \int_V D_{i,i} \delta_\varphi \varphi dV + \int_a n_i D_i \varphi_{,j} \delta u_j da \\
&\quad - \int_V \left(\varphi_{,j} D_i \delta u_{j,i} + \varphi_{,j} D_{i,i} \delta u_j \right) dV \\
&= \int_a n_i D_i \delta_\varphi \varphi da - \int_V D_{i,i} \delta_\varphi \varphi dV + \int_a n_i D_i \varphi_{,j} \delta u_j da \\
&\quad - \int_a n_i D_i \varphi_{,j} \delta u_j da + \int_V \left(\varphi_{,j} D_i \right)_{,i} \delta u_j dV - \int_V \varphi_{,j} D_{i,i} \delta u_j dV \\
&= \int_a n_i D_i \delta \varphi da - \int_V D_{i,i} \delta \varphi dV - \int_a n_i D_i \varphi_{,j} \delta u_j da + \int_V \left(\varphi_{,j} D_i \right)_{,i} \delta u_j dV
\end{aligned} \tag{27}
$$

and

$$
\int_V Q_{ij} \delta \varphi_{,ij} dV = \int_V Q_{ij} \left(\delta_\varphi \varphi_{,ij} + \varphi_{,ikj} \delta u_k \right) dV \tag{28}
$$

The first term on the right hand side of equation (28) can further reduce to

$$
\begin{aligned}
\int_V Q_{ij} \delta_\varphi \varphi_{,ij} dV &= \int_a n_j Q_{ij} \delta_\varphi \varphi_{,i} da - \int_V Q_{ij,j} \delta_\varphi \varphi_{,i} dV \\
&= - \int_a n_i Q_{ij,j} \delta_\varphi \varphi da + \int_V Q_{ij,ji} \delta_\varphi \varphi dV + \int_a n_j Q_{ij} \delta_\varphi \varphi_{,i} da
\end{aligned} \tag{29}
$$

and the second term on the right hand side of equation (28) can reduce to

$$\begin{aligned}
\int_V Q_{ij}\varphi_{,ikj}\delta u_k dV &= \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da - \int_V Q_{ij,j}\varphi_{,ik}\delta u_k dV - \int_V Q_{ij}\varphi_{,ik}\delta u_{k,j} dV \\
&= \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da - \int_a n_i Q_{ij,j}\varphi_{,k}\delta u_k da + \int_V \varphi_{,k}\left(Q_{ij,j}\delta u_k\right)_{,i} dV \\
&\quad - \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da + \int_V \left(Q_{ij}\varphi_{,ik}\right)_{,j}\delta u_k dV \\
&= -\int_a n_i Q_{ij,j}\varphi_{,k}\delta u_k dV + \int_V \varphi_{,k} Q_{ij,ji}\delta u_k dV + \int_V \varphi_{,k} Q_{ij,j}\delta u_{k,i} dV \\
&\quad + \int_V \left(Q_{ij}\varphi_{,ik}\right)_{,j}\delta u_k dV + \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da - \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da \\
&= -\int_a n_i Q_{ij,j}\delta_u \varphi dV + \int_V Q_{ij,ji}\delta_u \varphi dV + \int_V \left(Q_{ij}\varphi_{,ik}\right)_{,j}\delta u_k dV \\
&\quad - \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da \\
&\quad + \int_a n_i \varphi_{,k} Q_{ij,j}\delta u_k da - \int_V \left(\varphi_{,k} Q_{ij,j}\right)_{,i}\delta u_k dV + \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da
\end{aligned} \tag{30}$$

The sum of the last term of equation (29) and the last term of equation (30) can be written as

$$\begin{aligned}
&\int_a n_j Q_{ij}\delta_\varphi \varphi_{,i} da + \int_a n_j Q_{ij}\varphi_{,ik}\delta u_k da \\
&= \int_a n_j Q_{ij}\delta_\varphi \varphi_{,i} da + \int_a n_j Q_{ij}\delta_u \varphi_{,i} da = \int_a n_j Q_{ij}\delta \varphi_{,i} da
\end{aligned} \tag{31}$$

Since $\delta u_{i,j}$ is not independent of δu_i, and $\delta\varphi_{,i}$ is not independent of $\delta\varphi$ on the surface a, in order to identify the independent traction and charge boundary conditions, we separate the integrals including $\delta u_{i,j}$ and the integrals including $\delta\varphi_{,i}$ respectively as

$$\tau_{ijm} n_m \delta u_{i,j} = \tau_{ijm} n_m \Delta_j \delta u_i + \tau_{ijm} n_m n_j \Delta \delta u_i \tag{32}$$

$$n_j Q_{ij}\delta\varphi_{,i} = n_j Q_{ij}\Delta_i \delta\varphi + n_j Q_{ij} n_i \Delta\delta\varphi \tag{33}$$

by decomposing the gradient $\delta u_{i,j}$ into a tangential gradient $\Delta_j \delta u_i$ and a normal gradient $n_j \Delta \delta u_i$, and decomposing $\delta \varphi_{,i}$ into a tangential gradient $\Delta_i \delta \varphi$ and a normal gradient $n_i \Delta \delta \varphi$, *viz*.,

$$\delta u_{i,j} = \Delta_j \delta u_i + n_j \Delta \delta u_i \tag{34}$$

$$\delta \varphi_{,i} = \Delta_j \delta \varphi + n_j \Delta \varphi \tag{35}$$

where the operators $\Delta_j \equiv \left(\delta_{jk} - n_j n_k\right) \partial_k \equiv \nabla_s$, $\Delta \equiv n_k \partial_k$, and ∂_k denotes the partial derivative with respect to x_k. The terms in (32) may be resolved, further, in more than one way. For the first terms on the right hand side of (32) and (33), which contains the non-independent variation $\tau_{ijm} n_m \Delta_j \delta u_i$ and $n_j Q_{ij} \Delta_i \delta \varphi$ respectively, and can be rewritten as (Mindlin, 1964)

$$\tau_{ijm} n_m \Delta_j \delta u_i = \Delta_j \left(\tau_{ijm} n_m \delta u_i\right) - n_m \Delta_j \tau_{ijm} \delta u_i - \Delta_j \left(n_m\right) \tau_{ijm} \delta u_i \tag{36}$$

$$n_j Q_{ij} \Delta_i \varphi = \Delta_i \left(n_j Q_{ij} \delta \varphi\right) - n_j \Delta_i \left(Q_{ij}\right) \delta \varphi - \Delta_i \left(n_j\right) Q_{ij} \delta \varphi \tag{37}$$

The last two terms in the right hand side of (36) and (37), now contain the independent variations δu_i and $\delta \varphi$, respectively. For the preceding terms of equations (36) and (37), we note that, on the surface a,

$$\Delta_j \left(\tau_{ijm} n_m \delta u_i\right) = \left(\Delta_l n_l\right) n_j n_m \tau_{ijm} \delta u_i + n_q e_{qpk} \partial_p \left(e_{klj} n_l n_m \tau_{ijm} \delta u_i\right) \tag{38}$$

$$\Delta_i \left(n_j Q_{ij} \delta \varphi\right) = \left(\Delta_l n_l\right) n_i n_j Q_{ij} \delta \varphi + n_q e_{qpn} \partial_p \left(e_{nli} n_l n_j Q_{ij} \delta \varphi\right) \tag{39}$$

where e_{qpk} is the alternating tensor. By Stokes's theorem, the integrals, over a smooth surface, of the last terms in (38) and (39) vanish.

Similarly, the second part of equation (22) can be reduced to

$$\begin{aligned} &\int_V \frac{1}{2} \left(\sigma_{ij} u_{i,j} + \tau_{ijm} u_{i,jm} - D_i E_i - Q_{ij} V_{ij}\right) \delta u_{k,k} dV \\ =&\frac{1}{2} \int_a \left(\sigma_{ij} u_{i,j} + \tau_{ijm} u_{i,jm} + D_i \varphi_{,i} + Q_{ij} \varphi_{,ij}\right) n_k \delta u_k da \\ &- \frac{1}{2} \int_V \left(\sigma_{ij} u_{i,j} + \tau_{ijm} u_{i,jm} + D_i \varphi_{,i} + Q_{ij} \varphi_{,ij}\right)_{,k} \delta u_k dV \end{aligned} \tag{40}$$

Thus, the variation of the electric Gibbs free energy can be obtained as

$$
\begin{aligned}
\delta \int_V U_b dV = & \int_a \sigma_{ij} n_j \delta u_i da - \int_V \sigma_{ij,j} \delta u_i dV + \int_a (\Delta_l n_l)\, \tau_{ijm} n_m n_j \delta u_i da \\
& - \int_a \Delta_j (\tau_{ijm} n_m)\, \delta u_i da + \int_a \tau_{ijm} n_m n_j \Delta \delta u_i da - \int_a \tau_{ijm,m} n_j \delta u_i da \\
& + \int_V \tau_{ijm,mj} \delta u_i dV + \int_a n_i D_i \delta \varphi da - \int_V D_{i,i} \delta \varphi dV - \int_a n_i D_i \varphi_{,j} \delta u_j da \\
& + \int_V (\varphi_{,j} D_i)_{,i} \delta u_j dV - \int_a n_i Q_{ij,j} \delta \varphi da \\
& + \int_V Q_{ij,ji} \delta \varphi dV + \int_V (Q_{ij} \varphi_{,ik})_{,j} \delta u_k dV - \int_a n_j Q_{ij} \varphi_{,ik} \delta u_k da \\
& + \int_a n_i \varphi_{,k} Q_{ij,j} \delta u_k da - \int_V (\varphi_{,k} Q_{ij,j})_{,i} \delta u_k dV + \int_a n_j Q_{ij} n_i \Delta \delta \varphi da \\
& - \int_a \Delta_i (n_j Q_{ij})\, \delta \varphi da + \int_a (\Delta_l n_l)\, n_i n_j Q_{ij} \delta \varphi da \\
& + \frac{1}{2} \int_a (\sigma_{ij} u_{i,j} + \tau_{ijm} u_{i,jm} + D_i \varphi_{,i} + Q_{ij} \varphi_{,ij})\, n_k \delta u_k da \\
& - \frac{1}{2} \int_V (\sigma_{ij} u_{i,j} + \tau_{ijm} u_{i,jm} + D_i \varphi_{,i} + Q_{ij} \varphi_{,ij})_{,k} \delta u_k dV
\end{aligned}
\tag{41}
$$

The above equation can be further rewritten as

$$
\begin{aligned}
\delta \int_V U_b dV = & \int_a \left(\sigma_{ij} - \tau_{ijm,m} + \sigma_{ij}^{ES}\right) n_j \delta u_i da - \int_V \left(\sigma_{ij} - \tau_{ijm,m} + \sigma_{ij}^{ES}\right)_{,j} \delta u_i dV \\
& + \int_a \left[(\Delta_l n_l)\, \tau_{ijm} n_m n_j - \Delta_j (\tau_{ijm} n_m)\right] \delta u_i da + \int_a \tau_{ijm} n_m n_j \Delta \delta u_i da \\
& + \int_a n_j Q_{ij} n_i \Delta \delta \varphi da + \int_a \left[(\Delta_l n_l)\, n_i n_j Q_{ij} - \Delta_i (n_j Q_{ij})\right] \delta \varphi da \\
& + \int_a n_i (D_i - Q_{ij,j})\, \delta \varphi da - \int_V (D_i - Q_{ij,j})_{,i} \delta \varphi dV
\end{aligned}
\tag{42}
$$

where σ_{ij}^{ES} is the generalized bulk electrostatic force, which is defined as

$$\sigma_{ij}^{ES} = -\varphi_{,i}D_j - Q_{kj}\varphi_{,ki} + \varphi_{,i}Q_{jk,k} + \frac{1}{2}\left(\sigma_{ij}u_{i,j} + \tau_{ijm}u_{i,jm} + D_k\varphi_{,k} + Q_{kl}\varphi_{,kl}\right)\delta_{ij} \tag{43}$$

Comparing with σ_{ij} and $\tau_{ijm,m}$ the terms $\sigma_{kl}u_{k,l}$ and $\tau_{klm}u_{k,lm}$ can be neglected since the strain is small. Hence, equation (43) can be reduced to

$$\sigma_{ij}^{ES} = \sigma_{ij}^{M} - \tau_{ijm,m}^{M} \tag{44}$$

where σ_{ij}^{M} and $\tau_{ijm,m}^{M}$ are the generalized bulk Maxwell stress and the generalized bulk electrostatic stress corresponding to the electric field gradient, respectively, and defined as

$$\sigma_{ij}^{M} = -\varphi_{,i}D_j + \frac{1}{2}D_k\varphi_{,k}\delta_{ij} \tag{45}$$

$$\tau_{ijm,m}^{M} = -Q_{kj}\varphi_{,ki} + \varphi_{,i}Q_{jk,k} + \frac{1}{2}Q_{kl}\varphi_{,kl}\delta_{ij} \tag{46}$$

Equation (42) can be rewritten in tensors as

$$\begin{aligned}
\delta\int_V U_b dV = &-\int_V \left[\nabla\cdot\left(\boldsymbol{\sigma} - \nabla\cdot\tau + \boldsymbol{\sigma}^{ES}\right)\right]\cdot\left(\delta\mathbf{u}_t + \delta\mathbf{u}_n\right)dV \\
&-\int_V \nabla\cdot\left(\mathbf{D} - \nabla\cdot\mathbf{Q}\right)\delta\varphi dV + \int_a \mathbf{T}\cdot\left(\delta\mathbf{u}_t + \delta\mathbf{u}_n\right)da \\
&+\int_a \boldsymbol{\tau}:\left(\mathbf{n}\otimes\mathbf{n}\right)\cdot\Delta\delta\mathbf{u}da + \int_a q\delta\varphi da + \int_a \mathbf{Q}:\left(\mathbf{n}\otimes\mathbf{n}\right)\Delta\delta\varphi da
\end{aligned} \tag{47}$$

with $\mathbf{T}$ and q represents the force and charge, respectively,

$$T_i = \left(\sigma_{ij} - \tau_{ijm,m} + \sigma_{ij}^{ES}\right)n_j + \left(\Delta_l n_l\right)\tau_{ijm}n_m n_j - \Delta_j\left(\tau_{ijm}n_m\right) \tag{48a}$$

$$q = \left(D_i - Q_{ij,j}\right)n_i + \left(\Delta_l n_l\right)n_i n_j Q_{ij} - \Delta_i\left(n_j Q_{ij}\right) \tag{48b}$$

Now, we turn to the variation of the surface electric Gibbs free energy. From equation (21), the variation of the surface electric Gibbs free energy can be written as

$$\begin{aligned}\delta\int_a U_s da =& \int_a \delta U_s da + \int_a U_s \delta\left(\nabla_s\cdot\mathbf{u}\right)da\\ =&\int_a \left(\boldsymbol{\sigma}_s : \delta\boldsymbol{\varepsilon}_s + \boldsymbol{\tau}_s\vdots\delta\mathbf{w}_s + \mathbf{D}_s\cdot\delta\nabla_s\varphi + \mathbf{Q}_s : \delta\left(\nabla_s\otimes\nabla_s\varphi\right)\right)da\\ &+\int_a \frac{1}{2}[2U_{s0} + (\boldsymbol{\sigma}_s+\boldsymbol{\Gamma}) : \boldsymbol{\varepsilon}_s + (\boldsymbol{\tau}_s+\boldsymbol{\omega})\vdots\mathbf{w}_s\\ &+(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi + (\mathbf{Q}_s+\boldsymbol{\kappa}) : (\nabla_s\otimes\nabla_s\varphi)]\delta\left(\nabla_s\cdot\mathbf{u}\right)da\end{aligned} \tag{49}$$

where

$$\nabla_s\cdot\mathbf{u} = \mathrm{div}_s\mathbf{u} = \mathrm{tr}\left(\nabla_s\otimes\mathbf{u}\right) \tag{50}$$

By means of $\nabla_s\otimes\mathbf{u} = \nabla_s\otimes\mathbf{u}_t - u^n\mathbf{k}$, the above equation can be written as

$$\nabla_s\cdot\mathbf{u} = \mathrm{div}_s\mathbf{u} = \nabla_s\cdot\mathbf{u}_t - 2\gamma u^n \tag{51}$$

where the mean curvature γ can be obtained as

$$\gamma = \frac{1}{2}\mathrm{tr}\mathbf{k} = \frac{1}{2}\mathbf{I} : \mathbf{k} \tag{52}$$

Analogous to the variation in bulk, on the surface we have the similar relations as

$$\delta\varphi = \delta_\varphi\varphi + \delta_u\varphi = \delta_\varphi\varphi + \nabla_s\varphi\cdot\delta\mathbf{u} \tag{53}$$

$$\delta\nabla_s\varphi = \delta_\varphi\nabla_s\varphi + \delta_u\nabla_s\varphi = \delta_\varphi\nabla_s\varphi + \nabla_s\otimes\nabla_s\varphi\cdot\delta\mathbf{u} \tag{54}$$

$$\begin{aligned}\delta\left(\nabla_s\otimes\nabla_s\varphi\right) =&\delta_\varphi\left(\nabla_s\otimes\nabla_s\varphi\right) + \delta_u\left(\nabla_s\otimes\nabla_s\varphi\right)\\ =&\delta_\varphi\left(\nabla_s\otimes\nabla_s\varphi\right) + \nabla_s\otimes\nabla_s\otimes\nabla_s\varphi\cdot\delta\mathbf{u}\end{aligned} \tag{55}$$

where on the surface, $\delta_\varphi\varphi$, $\delta_\varphi\nabla_s\varphi$ and $\delta\left(\nabla_s\otimes\nabla_s\varphi\right)$ are produced by the virtual electric potential, $\delta_u\varphi = \nabla_s\varphi\cdot\delta\mathbf{u}$, $\delta_u\nabla_s\varphi = \nabla_s\otimes\nabla_s\varphi\cdot\delta\mathbf{u}$, and $\delta_u\left(\nabla_s\otimes\nabla_s\varphi\right) = \nabla_s\otimes\nabla_s\otimes\nabla_s\varphi\cdot\delta\mathbf{u}$ are produced on the surface by the virtual displacement.

Consider a region enclosed by an arbitrary closed smooth curve c in the curved surface a. The integrals along the curve c over a smooth surface should be zero, hence are omitted in this paper. Using the Green-Stokes theorem, the first part of equation (49) can reduce to

$$
\begin{aligned}
&\int_a \delta U_s da \\
&\quad = \int_a \left[\boldsymbol{\sigma}_s : \delta\left(\nabla_s \otimes \mathbf{u}_t - u^n \mathbf{k}\right) + \boldsymbol{\tau}_s \vdots \delta\left(\nabla_s \otimes \nabla_s \otimes \mathbf{u}_t - \nabla_s \otimes \left(u^n \mathbf{k}\right)\right) \right] da \\
&\qquad + \int_a \left[\mathbf{D}_s \cdot \delta \nabla_s \varphi + \mathbf{Q}_s : \delta\left(\nabla_s \otimes \nabla_s \varphi\right) \right] da \\
&\quad = \int_a \left[\nabla_s \cdot \left(\boldsymbol{\sigma}_s \cdot \delta \mathbf{u}_t\right) - \left(\nabla_s \cdot \boldsymbol{\sigma}_s\right) \delta \mathbf{u}_t - \left(\boldsymbol{\sigma}_s : \mathbf{k}\right) \delta u^n + \nabla_s \cdot \left[\boldsymbol{\tau}_s : \delta\left(\nabla_s \otimes \mathbf{u}_t\right)\right] \right. \\
&\qquad \left. - \left(\nabla_s \cdot \boldsymbol{\tau}_s\right) : \delta\left(\nabla_s \otimes \mathbf{u}_t\right) - \nabla_s \cdot \left(\boldsymbol{\tau}_s : \mathbf{k} \delta u^n\right) + \left(\nabla_s \cdot \boldsymbol{\tau}_s\right) : \mathbf{k} \delta u^n \right] da \\
&\qquad + \int_a \left[\mathbf{D}_s \cdot \delta \nabla_s \varphi + \mathbf{Q}_s : \delta\left(\nabla_s \otimes \nabla_s \varphi\right) \right] da \\
&\quad = - \int_a \left[\left(\nabla_s \cdot \boldsymbol{\sigma}_s\right) \cdot \delta \mathbf{u}_t + \left(\boldsymbol{\sigma}_s : \mathbf{k}\right) \delta u^n \right] da + \int_a \nabla_s \cdot \left(\nabla_s \cdot \boldsymbol{\tau}_s\right) \cdot \delta \mathbf{u}_t da \\
&\qquad + \int_a \left(\nabla_s \cdot \boldsymbol{\tau}_s\right) : \mathbf{k} \delta u^n da + \int_a \left[\mathbf{D}_s \cdot \delta \nabla_s \varphi + \mathbf{Q}_s : \delta\left(\nabla_s \otimes \nabla_s \varphi\right) \right] da
\end{aligned}
\tag{56}
$$

The last two terms on the right hand side of the above equation can reduce, respectively, to

$$
\begin{aligned}
&\int_a \mathbf{D}_s \cdot \delta \nabla_s \varphi da \\
&= \int_a \left[\mathbf{D}_s \cdot \delta_\varphi \nabla_s \varphi + \mathbf{D}_s \cdot \left(\nabla_s \otimes \nabla_s \varphi\right) \cdot \delta \mathbf{u} \right] da \\
&= - \int_a \nabla_s \cdot \mathbf{D}_s \delta_\varphi \varphi da - \int_a \nabla_s \cdot \left(\delta \mathbf{u} \otimes \mathbf{D}_s\right) \cdot \nabla_s \varphi da \\
&= - \int_a \nabla_s \cdot \mathbf{D}_s \delta_\varphi \varphi da - \int_a \left[\left(\nabla_s \varphi \otimes \mathbf{D}_s\right) : \delta\left(\nabla_s \otimes \mathbf{u}\right) + \left(\nabla_s \cdot \mathbf{D}_s\right) \nabla_s \varphi \cdot \delta \mathbf{u} \right] da \\
&= - \int_a \nabla_s \cdot \mathbf{D}_s \delta_\varphi \varphi da \\
&\quad - \int_a \left[\left(\nabla_s \varphi \otimes \mathbf{D}_s\right) : \delta\left(\nabla_s \otimes \mathbf{u}_t - u^n \mathbf{k}\right) \cdot \mathbf{D}_s + \left(\nabla_s \cdot \mathbf{D}_s\right) \nabla_s \varphi \cdot \delta \mathbf{u} \right] da
\end{aligned}
$$

$$= -\int_a \nabla_s \cdot \mathbf{D}_s \delta_\varphi \varphi da + \int_a \nabla_s \cdot (\nabla_s \varphi \otimes \mathbf{D}_s) \cdot \delta \mathbf{u}_t da - \int_a (\nabla_s \cdot \mathbf{D}_s) \nabla_s \varphi \cdot \delta \mathbf{u} da$$
$$+ \int_a (\nabla_s \varphi \otimes \mathbf{D}_s) : \mathbf{k} \delta u^n da \tag{57}$$
$$= -\int_a \nabla_s \cdot \mathbf{D}_s \delta \varphi da + \int_a \nabla_s \cdot (\nabla_s \varphi \otimes \mathbf{D}_s) \cdot \delta \mathbf{u}_t da + \int_a (\nabla_s \varphi \otimes \mathbf{D}_s) : \mathbf{k} \delta u^n da$$

and

$$\int_a \mathbf{Q}_s : \delta (\nabla_s \otimes \nabla_s \varphi) da$$
$$= \int_a \mathbf{Q}_s : \delta_\varphi (\nabla_s \otimes \nabla_s \varphi) da + \int_a \mathbf{Q}_s : [(\nabla_s \otimes \nabla_s \otimes \nabla_s \varphi) \cdot \delta \mathbf{u}] da \tag{58}$$

The first term of the right hand side of the above equation can be reduced to

$$\int_a \mathbf{Q}_s : \delta_\varphi (\nabla_s \otimes \nabla_s \varphi) da = -\int_a \nabla_s \cdot \mathbf{Q}_s \cdot \delta_\varphi \nabla_s \varphi da$$
$$= \int_a \nabla_s \cdot \nabla_s \cdot \mathbf{Q}_s \delta_\varphi \varphi da \tag{59}$$

The last term of the right hand side of equation (58) can be reduced to

$$\int_a \mathbf{Q}_s : [(\nabla_s \otimes \nabla_s \otimes \nabla_s \varphi) \cdot \delta \mathbf{u}] da$$
$$= -\int_a \nabla_s \cdot \mathbf{Q}_s \cdot (\nabla_s \otimes \nabla_s \varphi) \cdot \delta \mathbf{u} da - \int_a [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] : (\nabla_s \otimes \delta \mathbf{u}) da$$
$$= -\int_a \nabla_s \cdot \mathbf{Q}_s \cdot (\nabla_s \otimes \nabla_s \varphi) \cdot \delta \mathbf{u} da - \int_a [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] : (\nabla_s \otimes \delta \mathbf{u}_t - \delta u^n \mathbf{k}) da$$

$$
\begin{aligned}
&= \int_a \nabla_s \cdot [\delta \mathbf{u} \otimes (\nabla_s \cdot \mathbf{Q}_s)] \cdot \nabla_s \varphi da + \int_a \nabla_s \cdot [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] \cdot \delta \mathbf{u}_t da \\
&\quad + \int_a [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] : \mathbf{k} \delta u^n da \\
&= \int_a (\nabla_s \cdot \nabla_s \cdot \mathbf{Q}_s) \nabla_s \varphi \cdot \delta \mathbf{u} da + \int_a [\nabla_s \varphi \otimes (\nabla_s \cdot \mathbf{Q}_s)] : (\nabla_s \otimes \delta \mathbf{u}) da \\
&\quad + \int_a \nabla_s \cdot [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] \cdot \delta \mathbf{u}_t da + \int_a [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] : \mathbf{k} \delta u^n da \\
&= \int_a (\nabla_s \cdot \nabla_s \cdot \mathbf{Q}_s) \delta_u \varphi da + \int_a \nabla_s \cdot [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] \cdot \delta \mathbf{u}_t da \\
&\quad + \int_a [(\nabla_s \otimes \nabla_s \varphi) \cdot \mathbf{Q}_s] : \mathbf{k} \delta u^n da - \int_a \nabla_s \cdot [\nabla_s \varphi \otimes (\nabla_s \cdot \mathbf{Q}_s)] \cdot \delta \mathbf{u}_t da \\
&\quad - \int_a [\nabla_s \varphi \otimes (\nabla_s \cdot \mathbf{Q}_s)] : \mathbf{k} \delta u^n da
\end{aligned}
\tag{60}
$$

The variation of the second term on the right hand side of equation (49) can be written as

$$
\begin{aligned}
&\int_a U_s \delta (\nabla_s \cdot \mathbf{u}) da \\
&= \int_a \frac{1}{2} [2U_{s0} + (\boldsymbol{\sigma}_s + \boldsymbol{\Gamma}) : \boldsymbol{\varepsilon}_s + (\boldsymbol{\tau}_s + \boldsymbol{\omega}) \vdots \mathbf{w}_s \\
&+ (\mathbf{D}_s + \boldsymbol{\omega}) \cdot \nabla_s \varphi + (\mathbf{Q}_s + \boldsymbol{\kappa}) : (\nabla_s \otimes \nabla_s \varphi)] \delta (\nabla_s \cdot \mathbf{u}_t - 2\gamma u_n) da \\
&= - \int_a \{ \frac{1}{2} \nabla_s [2U_{s0} + (\sigma_s + \boldsymbol{\Gamma}) : \boldsymbol{\varepsilon}_s + (\boldsymbol{\tau}_s + \boldsymbol{\omega}) \vdots \mathbf{w}_s + (\mathbf{D}_s + \boldsymbol{\omega}) \cdot \nabla_s \varphi \\
&+ (\mathbf{Q}_s + \boldsymbol{\kappa}) : (\nabla_s \otimes \nabla_s \varphi)] \delta \mathbf{u}_t + [2U_{s0} + (\boldsymbol{\sigma}_s + \boldsymbol{\Gamma}) : \boldsymbol{\varepsilon}_s + (\boldsymbol{\tau}_s + \boldsymbol{\omega}) \vdots \mathbf{w}_s \\
&+ (\mathbf{D}_s + \boldsymbol{\omega}) \cdot \nabla_s \varphi + (\mathbf{Q}_s + \boldsymbol{\kappa}) : (\nabla_s \otimes \nabla_s \varphi)] \gamma \delta u_n \} da
\end{aligned}
\tag{61}
$$

Then, by substituting equations (56)-(61) into equation (49), we have

$$
\begin{aligned}
\delta\int_a U_s da = & \int_a [-\nabla_s\cdot(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s)]\,\delta\mathbf{u}_t da-\int_a [(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s):\mathbf{k}]\,\delta u^n da \\
& -\int_a \{\frac{1}{2}\nabla_s[2U_{s0}+(\sigma_s+\boldsymbol{\Gamma}):\boldsymbol{\varepsilon}_s+(\boldsymbol{\tau}_s+\boldsymbol{\omega})\vdots\mathbf{w}_s+(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi \\
& +(\mathbf{Q}_s+\boldsymbol{\kappa}):(\nabla_s\otimes\nabla_s\varphi)]\delta\mathbf{u}_t+[2U_{s0}+(\boldsymbol{\sigma}_s+\boldsymbol{\Gamma}):\boldsymbol{\varepsilon}_s+(\boldsymbol{\tau}_s+\boldsymbol{\omega})\vdots\mathbf{w}_s \\
& +(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi+(\mathbf{Q}_s+\boldsymbol{\kappa}):(\nabla_s\otimes\nabla_s\varphi)]\gamma\delta u_n\}da \\
& +\int_a \nabla_s\cdot[(\nabla_s\varphi\otimes\mathbf{D}_s)+(\nabla_s\otimes\nabla_s\varphi)\cdot\mathbf{Q}_s-\nabla_s\varphi\otimes(\nabla_s\cdot\mathbf{Q}_s)]\cdot\delta\mathbf{u}_t da \\
& +\int_a [(\nabla_s\varphi\otimes\mathbf{D}_s)+(\nabla_s\otimes\nabla_s\varphi)\cdot\mathbf{Q}_s-\nabla_s\varphi\otimes(\nabla_s\cdot\mathbf{Q}_s)]:\mathbf{k}\delta u^n da \\
& -\int_a \nabla_s\cdot(\mathbf{D}_s-\nabla_s\cdot\mathbf{Q}_s)\,\delta\varphi da
\end{aligned}
\tag{62}
$$

The above equation can be further rewritten as

$$
\begin{aligned}
\delta\int_a U_s da = & \int_a \left[-\nabla_s\cdot\left(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s+\boldsymbol{\sigma}_s^{ES}\right)\right]\delta\mathbf{u}_t da \\
& +\int_a \left[-\left(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s+\boldsymbol{\sigma}_s^{ES}\right):\mathbf{k}\right]\delta u^n da-\int_a \nabla_s\cdot(\mathbf{D}_s-\nabla_s\cdot\mathbf{Q}_s)\,\delta\varphi da
\end{aligned}
\tag{63}
$$

where $\boldsymbol{\sigma}_s^{ES}$ is the generalized surface electrostatic force, which is defined as

$$
\begin{aligned}
\boldsymbol{\sigma}_s^{ES} = & -(\nabla_s\varphi\otimes\mathbf{D}_s)-(\nabla_s\otimes\nabla_s\varphi)\cdot\mathbf{Q}_s+\nabla_s\varphi\otimes(\nabla_s\cdot\mathbf{Q}_s) \\
& +\frac{1}{2}[2U_{s0}+(\boldsymbol{\sigma}_s+\boldsymbol{\Gamma}):\boldsymbol{\varepsilon}_s+(\boldsymbol{\tau}_s+\boldsymbol{\omega})\vdots\mathbf{w}_s \\
& +(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi+(\mathbf{Q}_s+\boldsymbol{\kappa}):(\nabla_s\otimes\nabla_s\varphi)]\mathbf{I}
\end{aligned}
\tag{64}
$$

The expression (64) of the surface electrostatic force is almost same as the one for the bulk, *i.e.*, equation (43). Comparing with $\boldsymbol{\sigma}_s$ and $\nabla_s\cdot\boldsymbol{\tau}_s$ the term $\boldsymbol{\sigma}_s:\boldsymbol{\varepsilon}_s$ and $\boldsymbol{\tau}_s\vdots\mathbf{w}_s$ can be neglected since the strain is small. Hence, equation (64) can be reduced to

$$\begin{aligned}\boldsymbol{\sigma}_s^{ES} =& -(\nabla_s\varphi\otimes\mathbf{D}_s)-(\nabla_s\otimes\nabla_s\varphi)\cdot\mathbf{Q}_s+\nabla_s\varphi\otimes(\nabla_s\cdot\mathbf{Q}_s)\\ &+\frac{1}{2}\left[2U_{s0}+(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi+(\mathbf{Q}_s+\boldsymbol{\kappa}):(\nabla_s\otimes\nabla_s\varphi)\right]\mathbf{I}\\ =&\boldsymbol{\sigma}_s^M-\nabla_s\cdot\boldsymbol{\tau}_s^M\end{aligned} \tag{65}$$

where $\boldsymbol{\sigma}_s^M$ and $\nabla_s\cdot\boldsymbol{\tau}_s^M$ are the generalized surface Maxwell stress, and the generalized surface electrostatic stress corresponding to the electric field gradient effect, respectively, which are defined as

$$\boldsymbol{\sigma}_s^M=-(\nabla_s\varphi\otimes\mathbf{D}_s)+\frac{1}{2}\left[2U_{s0}+(\mathbf{D}_s+\boldsymbol{\omega})\cdot\nabla_s\varphi\right]\mathbf{I} \tag{66}$$

$$\begin{aligned}\nabla_s\cdot\boldsymbol{\tau}_s^M =& -(\nabla_s\otimes\nabla_s\varphi)\cdot\mathbf{Q}_s+\nabla_s\varphi\otimes(\nabla_s\cdot\mathbf{Q}_s)\\ &+\frac{1}{2}\left[(\mathbf{Q}_s+\boldsymbol{\kappa}):(\nabla_s\otimes\nabla_s\varphi)\right]\mathbf{I}\end{aligned} \tag{67}$$

Due to the arbitrariness of $\delta\mathbf{u}_t$, δu^n, and $\delta\varphi$, from equations (47) and (63), the governing equations can be written as

$$\nabla\cdot\left(\boldsymbol{\sigma}-\nabla\cdot\boldsymbol{\tau}+\boldsymbol{\sigma}^{ES}\right)=0 \tag{68}$$

$$\nabla\cdot(\mathbf{D}-\nabla\cdot\mathbf{Q})=0 \tag{69}$$

with the boundary conditions on a

$$\mathbf{T}\cdot\mathbf{R}=\nabla_s\cdot\left(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s+\boldsymbol{\sigma}_s^{ES}\right) \tag{70}$$

$$\mathbf{T}\cdot\mathbf{n}=\left(\boldsymbol{\sigma}_s-\nabla_s\cdot\boldsymbol{\tau}_s+\boldsymbol{\sigma}_s^{ES}\right):\mathbf{k} \tag{71}$$

$$q=\nabla_s\cdot(\mathbf{D}_s-\nabla_s\cdot\mathbf{Q}_s) \tag{72}$$

$$\boldsymbol{\tau}:(\mathbf{n}\otimes\mathbf{n})=0 \tag{73}$$

$$\mathbf{Q}:(\mathbf{n}\otimes\mathbf{n})=0 \tag{74}$$

Equations (70)-(72) become the generalized electromechanical Young-Laplace equations, where equation (72) is the electric Young-Laplace equation and represents the surface charge equation (surface Gauss's law). Equations (68) and (69), with boundary conditions (70)-(74), form the equations for nanosized linear elastic dielectrics with the surface and strain/electric field gradient effects, as well as the effect of the electrostatic force. After appropriate simplifications and transformations, these equations can be reduced to some special theory and those available in the literature. For example, without the surface effect, the strain gradient and the electric field gradient effect, these equations are reduced to the classical electromechanical problems (Yang, 2004a; Zhang, 2004). If we do not consider the electromechanical coupling and electric field gradient, only consider the electrostatics, equation (72) can reduce to the same as that in Slavchov *et al.* (2006) for surface polarization.

If we do not consider the effects of the electric field gradient and strain gradient, $\nabla \cdot \boldsymbol{\tau}^M$ and $\nabla_s \cdot \boldsymbol{\tau}_s^M$ should disappear. Then, without the external traction, the Young-Laplace equations become

$$\mathbf{n} \cdot \left(\boldsymbol{\sigma} + \boldsymbol{\sigma}^M\right) \cdot \mathbf{R} = \nabla_s \cdot \left(\boldsymbol{\sigma}_s + \boldsymbol{\sigma}_s^M\right) \tag{75}$$

$$\mathbf{n} \cdot \left(\boldsymbol{\sigma} + \boldsymbol{\sigma}^M\right) \cdot \mathbf{n} = \left(\boldsymbol{\sigma}_s + \boldsymbol{\sigma}_s^M\right) : \mathbf{k} \tag{76}$$

If the electrostatic force, strain gradient and the surface effect are not taken into account, then the governing equations reduce to those for electric field gradient in Yang (2004a, b). If the electrostatic force, strain gradient and the electric field gradient effects are not taken into account, we can obtain the surface piezoelectric theory.

If the electrostatic force are omitted, the second integrals in equations (20) and (21) can be neglected, then the electromechanical surface/gradient theory can be derived directly from (68)-(74) by leaving the generalized bulk and surface electrostatic stresses $\boldsymbol{\sigma}^{ES}$ and $\boldsymbol{\sigma}_s^{ES}$ out.

If the surface polarizations are neglected, the surface constitutive relations are pure mechanical, and the surface electrostatic stress $\boldsymbol{\sigma}_s^{ES}$ should be canceled from equations (70) and (71) while the right hand side of equation (72) becomes zero.

If we do not consider the electromechanical coupling, then we can obtain the surface/gradient elastic theory. Further, equations (68) and (69) will reduce to the classical surface elastic theory as in Gurtin (1975) if we do not consider the strain gradient effect either.

4 Conclusions

In this paper, a variational principle was established for nanosized dielectrics with the strain/electric field gradients and surface effects. Based on the variational principle, we formulated a theoretical framework to examine the size effect due to both the electric field gradient effect and surface effect for dielectrics. The surface effect includes both the effect of surface stress and surface polarization. The governing equations and the generalized electromechanical Young-Laplace equations, which include the effects of strain/electric field gradients, surface and electrostatic force, were presented. The generalized bulk and surface electrostatic stress are obtained from the variational principle naturally. These formulae are different from those for the flexoelectricity (Shen and Hu, 2010).

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