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Electro-elastic fields due to a point charge in a flexoelectric medium

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ABSTRACT

Flexoelectricity provides a two-way connection between strain gradients and polarization that is pronounced at the nanoscale for isotropic materials which cannot link electromechanically via piezoelectricity. In this paper, the general equations for an isotropic, flexoelectric material were formulated, with contributions from strain gradients included. The electromechanical fields associated with a point charge in an infinite medium were derived, and results for GaAs were obtained. Our formulation yields two electromechanical length-scales, instead of one obtained from previous theories, and enables us to capture local fields accurately. Results from this paper provide insight into the electro-mechanical behavior of materials with charged defects.

1. Introduction

Flexoelectricity is a type of electro-mechanical coupling mechanism that provides a linkage between mechanical strain gradient and electric polarization, as well as a connection between polarization gradient and mechanical strains. In analogy with piezoelectricity, the former is referred to as the direct flexoelectric effect, while the latter is indicated by the term "converse flexoelectric effect". From a macroscopic perspective, the salient feature that distinguishes flexo- and piezo-electricity is that flexoelectricity is a gradient phenomenon that can be significant only when strains and polarization are non-uniform. Further, since the material constants that characterize flexoelectricity are typically very small, flexoelectric contributions to polarization become appreciable only at small length scales due to the presence of high strain gradients associated with heterogeneities that may have high elastic and dielectric contrast with the parent material. Microscopically speaking, piezoelectricity occurs only in non-centrosymmetric materials (such as ZnO or any non-cubic phase of $BaTiO_3$) due to the creation of dipole moments upon application of strain. In contrast, flexoelectricity most clearly manifests itself in centrosymmetric materials (such as NaCl or cubic $BaTiO_3$) for which dipoles can only be created by applying a non-uniform strain so that the respective centroids of positive and negative charges shift away from each other [1]. In what follows, we will confine the discussion to flexoelectricity; for fundamentals of piezoelectricity and related boundary value problems, Ikeda [2] and the research of Pan and coworkers, e.g. [3–5] can be cited respectively.

Flexoelectricity has been a subject of considerable interest and has been studied both theoretically and experimentally – in various contexts over the last several decades. Given the availability of some excellent reviews [6–9], only a condensed review is provided here. In an early landmark paper on converse flexoelectricity, Mindlin [10] developed a continuum model that incorporated polarization gradient into the stored energy of elastic dielectrics. Mindlin's theory provided a mathematical basis for including the core–shell and shell–shell interactions between atoms, in addition to the dominant core–core interactions described by classical piezoelectricity. In a subsequent publication, Mindlin [11] successfully used his polarization gradient theory to explain the anomalous reduction in the capacitance of very thin dielectric plates. Further, Mindlin [12] showed that for rigid dielectrics, the classical Coulomb potential due to a point charge has to be augmented by an additional exponential term that decays at "large" distances from the point charge, but can provide a significant correction in its vicinity. An important limitation to Mindlin's work was that it did not account for the direct flexoelectric effect, i.e., the coupling between strain gradients and polarization. Askar et al. [13–15] used lattice dynamics models to determine the material properties for Mindlin's theory for certain crystals (e.g., NaCl and KCl) and provided solutions to boundary value problems involving cylindrical/spherical cavities and cracks. Chowdhury and Glockner [16] used Mindlin's polarization gradient theory to evaluate the fields associated with a point charge inside an elastic, dielectric half-space. Nowacki and coworkers [17,18] developed Green's functions for Mindlin's theory, and provided illustrative solutions to several one dimensional problems. A notable enhancement of Mindlin's work was the incorporation of the direct flexoelectric effect in the stored energy of a dielectric [7,1]. Maranganti and Sharma [7] derived Green's

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functions associated with a point force and a point electric field, and provided solutions to the corresponding Eshelby's inclusion problem [19]. Sharma et al. [1] explored the technologically-relevant "paradox" of creating "piezoelectric" dielectric nanocomposites without using piezoelectric constituents through the universal mechanism of flexoelectricity. One of the key requirements for such a material would be a net non-zero dipole moment in a representative volume element; in essence for a particulate nano-composite, this would necessitate the use of low-symmetry inclusion shapes that are oriented favorably in a particular direction. Recently, Shen and Hu [20] proposed a variational principle that accounts for flexoelectricity and surface effects for elastic dielectrics. More recently, Mao and Purohit [21] contributed solutions to some boundary value problems for flexoelectric solids. Morozovska and coworkers [22] used phase-field modeling and DFT computations to accentuate the role of flexoelectricity in defining the internal structure of ferroelectric domain walls. In another work, the authors [23] employed the Landau–Ginzburg–Devonshire theory to assess the role of flexoelectricity in the polarization of various types of interfaces in several otherwise non-ferroelectric perovskites such as $SrTiO_3$.

Despite the challenges associated with conducting experiments at small length scales where the flexoelectric effect becomes appreciable, numerous authors have reported findings consistent with this phenomenon. Electromechanical coupling in dislocated non-piezoelectric dielectric crystals was attributed to polarization in the vicinity of dislocations where high strain gradients are expected [24–27]. As already noted, Mindlin [11] accounted for the anomalously low capacitance of thin dielectric plates. Flexoelectricity has also been observed in the bending of thin, crystal plates [28] and in the inhomogeneous stretching of BST thin films [29]. The effect of curvature on the polarization of carbon nano-shells has been studied by Dumitrica et al. [30]. Cross and coworkers [31,32] have relied on flexoelectricity to engineer piezoelectric composites by using non-piezoelectric constituents.

Given that flexoelectricity is a higher-order effect in comparison with piezoelectricity, it is not surprising that the flexoelectric coefficients for most materials are fairly small. Indeed, Kogan [33] indicated that the lower bound for flexoelectric coefficients should be of the order of e/a (10^{-9} C/m), where e is the electronic charge and a is a lattice parameter. Similar estimates were derived by Marvan and Havranek [34,35] for glassy polymers and elastomers with a being interpreted to be a characteristic inter-atomic distance. A noteworthy feature of the elastomer model by these authors was the linear proportionality of the flexoelectric coefficient with the dielectric constant; this suggests that high flexoelectric coefficients can be found in materials that possess high dielectric constants. The experimental work of Cross and co-workers [36–39] has indicated high flexoelectric coefficients (10^{-6} C/m) in certain ferroelectric perovskites such as BST, PZT and PMN. High flexoelectric coefficients have also been reported for the polymer PVDF by Baskaran et al. [40,41]. However, their findings have been disputed by Chu and Salem [42], who have measured much lower flexoelectric coefficients for PVDF. The source of the disparate experimental findings for PVDF is probably in part due to the complexity of fully characterizing the microstructure of a polymer and how it relates to the observed flexoelectric effect.

In this paper, we revisit Mindlin's [12] solution to the problem of a point charge in a rigid dielectric in the context of an isotropic, flexoelectric material that includes energy terms from elastic strain-gradients. This paper will provide insight into the role of small charged defects on the electro-mechanical fields induced in the bulk of the material.

This paper is organized as follows. In Section 2, we outline the governing equations for isotropic, flexoelectric materials. In Section 3, we develop the solution for the electro-elastic fields associated with a point charge embedded in an infinite dielectric medium. This is followed by a discussion of salient results in Section 4. We conclude in Section 5.

2. Formulation

We begin by considering a dielectric material occupying volume Ω and bounded by surface $\partial\Omega$. The volume over all space is denoted as Ω^∞. The static version of Hamilton's principle states

$$-\delta\int_{\Omega^\infty} H dV + \int_\Omega \left(F_i\delta u_i + E_i^0\delta P_i - \rho\delta\phi\right) dV + \int_{\partial\Omega} t_i\delta u_i dS = 0 \tag{1}$$

where H is the electric enthalpy, F_i, E_i^0 and t_i are the applied body force, electric field, and traction respectively, ρ is the charge density, ϕ is the electric potential, and u_i and P_i are respectively the displacement and polarization fields. The electric enthalpy is taken to depend on the infinitesimal strain $\varepsilon_{ij} = (u_{i,j} + u_{j,i})/2$, P_i and their gradients, and the potential:

$$H = W^L(P_i, P_{i,j}, \varepsilon_{ij}, u_{i,jk}) - \tfrac{1}{2}\epsilon_0\phi_{,i}\phi_{,i} + \phi_{,i}P_i \tag{2}$$

where W^L is the energy density associated with the dielectric. The variation of H yields

$$\delta H = (-E_i + \phi_{,i})\delta P_i + Q_{ij}\delta P_{i,j} + \sigma_{ij}\delta\varepsilon_{ij} + T_{ijk}\delta u_{i,jk} + (P_i - \epsilon_0\phi_{,i})\delta\phi_{,i} \tag{3}$$

where the work conjugates $E_i \equiv -\partial W^L/\partial P_i$ and $Q_{ij} \equiv \partial W^L/\partial P_{i,j}$ are the electric field and higher-order electric force respectively, while $\sigma_{ij} \equiv \partial W^L/\partial\varepsilon_{ij}$ and $T_{ijk} \equiv \partial W^L/\partial u_{i,jk}$ are respectively the symmetric stress and a higher-order stress. Standard variational analysis can be used to derive the equilibrium equations, constitutive equations and the boundary conditions:

Equilibrium equations

$$\begin{aligned}
&\sigma_{ij,j} - T_{ijk,jk} + F_i = 0 \quad (\text{in } \Omega)\\
&E_i - \phi_{,i} + Q_{ij,j} + E_i^0 = 0 \quad (\text{in } \Omega)\\
&P_{i,i} - \epsilon_0\phi_{,ii} - \rho = 0 \quad (\text{in } \Omega)\\
&\phi_{,ii} = 0 \quad (\text{outside } \Omega)
\end{aligned} \tag{4}$$

Boundary conditions

$$\begin{aligned}
&n_j(\sigma_{ij} - T_{ijk,k}) - D_j^S(n_kT_{ijk}) + D^N(n_jn_kT_{ijk}) = t_i \quad (\text{on } \partial\Omega)\\
&T_{ijk}n_jn_k = 0 \quad (\text{on } \partial\Omega)\\
&\left[P_i + \epsilon_0\|\phi_{,i}\|\right]n_i = 0 \quad (\text{on } \partial\Omega)\\
&Q_{ij}n_j = 0 \quad (\text{on } \partial\Omega)
\end{aligned} \tag{5}$$

where $\|\phi_{,i}\|$ is the jump in the potential gradient across $\partial\Omega$. $D^N \equiv n_i\partial_i$ and $D_i^S \equiv \partial_i - n_iD^N$ are the normal and surface gradient operators respectively [43–45,21], and n_i is the unit normal vector to the surface $\partial\Omega$. In order to establish the constitutive equations, we consider the following form of the energy density [46,1]:

$$W^L = \tfrac{1}{2}a_{ij}P_iP_j + \tfrac{1}{2}b_{ijkl}P_{i,j}P_{k,l} + \tfrac{1}{2}c_{ijkl}\varepsilon_{ij}\varepsilon_{kl} + d_{ijkl}P_{i,j}\epsilon_{kl} + f_{ijkl}P_iu_{j,kl}$$

$$+ \tfrac{1}{2}g_{ijklmn}u_{i,jk}u_{l,mn} \tag{6}$$

where **a** is the reciprocal dielectric susceptibility tensor, **b** is the fourth order tensor that models energy contribution from polarization gradients, **c** is the linear elastic stiffness tensor, **d** is a tensor that links polarization gradient to strains (converse flexoelectric effect), **f** is a tensor that links polarization to strain gradient (direct