Flexoelectricity in soft materials and biological membranes

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ABSTRACT

Flexoelectricity and the concomitant emergence of electromechanical size-effects at the nanoscale have been recently exploited to propose tantalizing concepts such as the creation of "apparently piezoelectric" materials without piezoelectric materials, e.g. graphene, emergence of "giant" piezoelectricity at the nanoscale, enhanced energy harvesting, among others. The aforementioned developments pertain primarily to hard ceramic crystals. In this work, we develop a nonlinear theoretical framework for flexoelectricity in soft materials. Using the concept of soft electret materials, we illustrate an interesting nonlinear interplay between the so-called Maxwell stress effect and flexoelectricity, and propose the design of a novel class of apparently piezoelectric materials whose constituents are intrinsically non-piezoelectric. In particular, we show that the electret-Maxwell stress based mechanism can be combined with flexoelectricity to achieve unprecedentedly high values of electromechanical coupling. Flexoelectricity is also important for a special class of soft materials: biological membranes. In this context, flexoelectricity manifests itself as the development of polarization upon changes in curvature. Flexoelectricity is found to be important in a number of biological functions including hearing, ion transport and in some situations where mechanotransduction is necessary. In this work, we present a simple linearized theory of flexoelectricity in biological membranes and some illustrative examples.

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1. Introduction

Piezoelectrics are an intriguing class of materials where a uniform mechanical strain can induce an electric field and conversely, a uniform electric field can cause mechanical actuation. This phenomenon has found wide applications: in energy harvesting, sensing and actuation, advanced microscopes, artificial muscles, minimally invasive surgery among others (Wang et al., 2010; Madden et al., 2004; Gautschi, 2002; Labanca et al., 2008). Both soft materials (e.g. polymers) and hard crystalline ceramics exhibit this phenomenon albeit the microscopic mechanisms underpinning piezoelectricity differ in these two classes of materials (Furukawa, 1989; Damjanovic, 1998).

Recently, a somewhat understudied electromechanical coupling, flexoelectricity, has attracted a fair amount of attention from both fundamental and applications points of view leading to intensive experimental (Cross, 2006; Ma and Cross, 2001, 2002, 2003, 2006; Catalan et al., 2004; Zubko et al., 2007; Fu et al., 2006, 2007) and theoretical activity in this topic (Sharma

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et al., 2007; Eliseev et al., 2009, 2011; Maranganti and Sharma, 2009; Majdoub et al., 2008a,b, 2009a,b; Sharma et al., 2010, 2012; Gharbi et al., 2011; Kalinin and Meunier, 2008; Dumitrica et al., 2002). The aforementioned works and nearly all the current literature on flexoelectricity focus on crystalline materials. Since several concepts pertaining to flexoelectricity in crystalline materials also carry over to soft matter, in the following, we provide some discussion of the former subject also.

To understand flexoelectricity better, it is best first to allude to the central mathematical relation that describes piezoelectricity:

$$P_i \sim d_{ijk}\varepsilon_{jk} \tag{1}$$

In the above equation the polarization vector P_i is related to the second order strain tensor ε_{jk} through the third order piezoelectric material property tensor d_{ijk}. Tensor transformation properties require that under inversion-center symmetry, all odd-order tensors vanish. Thus, most common crystalline materials, e.g. Silicon, and NaCl are not piezoelectric whereas ZnO and GaAs are. Physically, however, it is possible to visualize how a non-uniform strain or the presence of strain gradients may potentially break the inversion symmetry and induce polarization even in centrosymmetric crystals (Tagantsev et al., 2009; Tagantsev, 1986; Maranganti et al., 2006). This is tantamount to extending relation (1) to include strain gradients:

$$P_i \sim d_{ijk}\varepsilon_{jk} + f_{ijkl}\frac{d\varepsilon_{jk}}{dx_l} \tag{2}$$

Here f_{ijkl} are the components of the so-called flexoelectric tensor. While the piezoelectric property is non-zero only for select materials, the strain gradient-polarization coupling (i.e., flexoelectricity tensor) is in principle non-zero for all (insulating) materials. This implies that under a non-uniform strain, all dielectric materials are capable of producing a polarization. The flexoelectric mechanism is well-illustrated by the non-uniform straining of a graphene nanoribbon—a manifestly non-piezoelectric material (Fig. 1(a)) (Dumitrica et al., 2002; Chandratre and Sharma, 2012). Flexoelectricity has been experimentally confirmed in several crystalline materials such as NaCl, ferroelectrics like Barium Titanate among others, e.g. Refs. (Fu et al., 2006, 2007). The mechanisms of flexoelectricity in polymers (while experimentally proven) still remain unclear (Baskaran et al., 2011, 2012; Chu and Salem, 2012) and atomistic modeling (being conducted by the authors) is expected to shed light on this issue in the near future. We speculate that the presence of frozen dipoles and their thermal fluctuations is the cause of flexoelectricity in soft materials, however, we cannot offer a more definitive explanation at this point and simply emphasize that this phenomenon has been experimentally confirmed (Baskaran et al., 2011, 2012; Chu and Salem, 2012) and further elucidation is a subject of future research. It is worthwhile to note that the term "flexoelectricity" was first coined in the context of liquid crystals (Meyer, 1969; deGennes, 1974). A substantial literature on flexoelectricity in thermotropic liquid crystals does exist—a detailed discussion of which is beyond the scope of the current paper. The reader is simply referred to a recent text that summarizes much of the literature on that topic (Eber and Buka, 2012).

Flexoelectricity results in the size-dependency of electromechanical coupling and researchers (including us) have advocated several tantalizing applications that can result through its exploitation. For example, the notion of creating piezoelectric materials without using piezoelectric materials (Sharma et al., 2007, 2010; Chandratre and Sharma, 2012; Fu et al., 2007), giant piezoelectricity in inhomogeneously deformed nanostructures (Majdoub et al., 2008a, 2009a), enhanced energy harvesting (Majdoub et al., 2008b, 2009b), the origins of nanoindentation size effects (Gharbi et al., 2011), renormalized ferroelectric properties (Catalan et al., 2004; Eliseev et al., 2009, 2011), the origins of the dead-layer effect in nanocapacitors (Majdoub et al., 2009) among others. In fact, Chandratre and Sharma (2012) have recently shown that graphene can be coaxed to behave like a piezoelectric material merely by creating holes of certain symmetry. The artificial piezoelectricity thus produced was found to be almost as strong as that of well-known piezoelectric substances such as quartz. Such a constructed graphene nanoribbon may be considered to be the thinnest known piezoelectric material. We briefly elaborate on this notion (Fig. 2). Consider a material consisting of two or more different non-piezoelectric dielectrics —as a concrete example that has been studied in the past we may think of a (dielectric) graphene nanoribbon impregnated
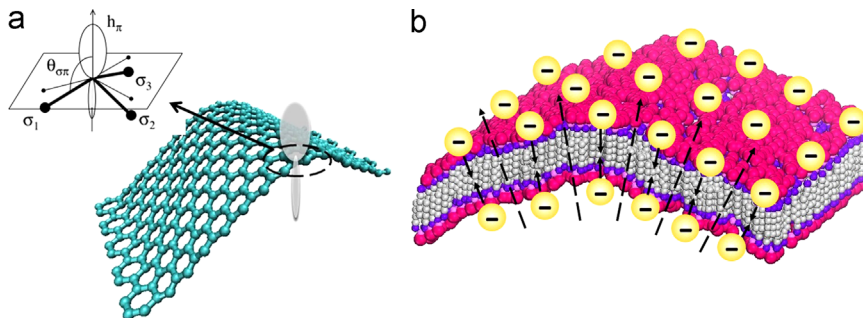

Fig. 1. Flexoelectricity in membranes. (a) Bending of graphene: upon bending, the symmetry of the electron distribution at each atomic site is broken, which leads to the polarization normal to the graphene ribbon; an infinite graphene sheet is semi-metallic; however, finite graphene nanoribbons can be dielectric depending upon surface termination. (b) Bending of a lipid bilayer membrane: due to bending, both the charge and dipole densities in the upper and lower layers become asymmetric. This asymmetry causes the normal polarization in the bilayer membrane.
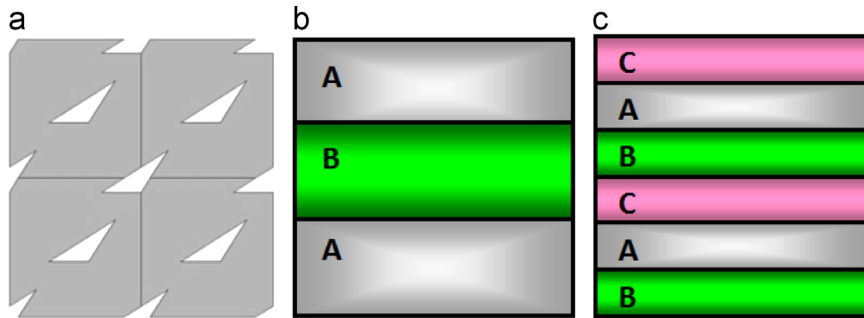
Fig. 2. Creating a piezoelectric material without using piezoelectric materials. (a) A material with a second phase, under a uniform stress, will produce local strain gradients and hence local polarization due to flexoelectricity. If the shape of the second phase is non-centrosymmetrical, the average polarization will be non-zero as well thus, from a macroscopic viewpoint, exhibiting a piezoelectric like effect. In this figure, this concept is illustrated by riddling a sheet with triangular holes. Such a sheet, with circular holes will yield a net zero average polarization. (b) The concept may also be realized by using a superlattice of differing materials. However, here care must be taken. A bilayer superlattice will result in a zero average polarization and (c) a trilayer superlattice is required to break the requisite symmetry. Nevertheless, finite bilayers (due to symmetry breaking at the free surfaces) will produce this effect.

with holes (Fig. 2(a)) (Chandratre and Sharma, 2012). Upon the application of uniform stress, differences in material properties at the interfaces of the materials will result in the presence of strain gradients. Those gradients will induce local spatially varying polarization due to the flexoelectric effect. As long as certain symmetry rules are followed, the net average polarization will be non-zero. Thus, the artificially structured material will exhibit an electrical response under uniform stress, behaving therefore like a piezoelectric material.[1] The length scales must be "small" since this concept requires very large strain gradients and those for a given strain are generated easily only at the nanoscale. Here we should mention that the precise scale at which this effect becomes prominent depends on the strength of the flexoelectric coefficients. For several materials studied in the past, sub-100 nm characteristic length scales are required (e.g. Majdoub et al., 2008a, 2009a; Eliseev et al., 2009) albeit (as this study will also show) the ramifications of flexoelectricity can also manifest with feature size of a few microns. Regarding symmetry: Topologies of only certain symmetries can realize the aforementioned concept. For example, circular holes distributed in a material will not yield apparent piezoelectric behavior even though the flexoelectric effect will cause local polarization fields. Due to circular symmetry, the overall average polarization is zero. A similar material, but containing triangular shaped holes or inclusions aligned along a given direction, will exhibit the required apparent piezoelectricity.[2] In a similar vein, a *finite* bilayer configuration may also be used although a bilayer *superlattice* would not work (Fig. 2(b))–see discussions in Sharma et al. (2010).

Flexoelectricity is also important for a special class of soft materials: biological membranes. In this context, flexoelectricity is referred to as the development of polarization upend changes in the curvature (Fig. 1(b)). Petrov has discussed this in a series of works (Petrov, 1975, 1998, 1999, 2002, 2006, 2007; Petrov et al., 1993, 1996) and references therein—these include several detailed experimental confirmation of this phenomenon. Simply, $\mathbf{P} = f_e K_h \mathbf{n}$. Here $\mathbf{P}$ is the polarization (per unit area), K_h is the mean curvature, $\mathbf{n}$ is the normal vector and f_e is the flexoelectric coupling constant that dictates the strength of the electromechanical coupling. A recent, quite definitive experimental evidence of flexoelectricity in biological membranes (specifically the converse effect) was recently provided in a work by Brownell et al. (2003) where its role in cell-membrane tether formation was also elucidated. In addition, flexoelectricity has been found to have important implications for ion transport (Petrov et al., 1993), hearing mechanism (Brownell et al., 2001, 2003; Raphael et al., 2000), tether formation (Breneman and Rabbitt, 2009) among others.

There are other types of electromechanical couplings. For example, all insulating materials exhibit electrostriction and the so-called "Maxwell stress effect" whereby an application of electric field can deform the material. Although mathematically similar, electrostriction and "Maxwell stress" have different physical origins (Zhao and Suo, 2008). In these two phenomena, however, a converse effect does not exist (in stark contrast to both flexoelectricity and piezoelectricity). Moreover, reversal of the electric field does not reverse the deformation direction. Maxwell stress effect and electrostriction, in addition to being only a one-way electromechanical coupling, are also inherently nonlinear in nature and usually only appreciable in soft materials. Indeed, there has been intense recent activity in understanding and exploiting electrostriction and Maxwell stress effect in soft materials. The motivation for considering soft electromechanical materials is well articulated by a recent overview article by Suo (2010). Inspired from nature, soft materials that respond to multi-field stimuli can be used in intriguing applications such as (more human like) soft robots and polymer actuators (Trivedi et al., 2008; Shankar et al., 2007; Pelrine et al., 2000). There is a fair amount of history associated with mechanics oriented research on this topic. We avoid an elaborate literature review but, to establish appropriate context, highlight a few papers that are pertinent to the present work. In particular, the reader is referred to Suo et al. (2008) for a literature survey. While one of the earliest definitive work on deformable electromechanical media is due to Toupin (1956), different flavors have appeared

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[1] The converse effect can also be shown to exist. A uniform electric field may induce a non-zero average strain.

[2] We remark that even for triangular holes or inclusions, if they are distributed randomly, the average polarization will be zero.
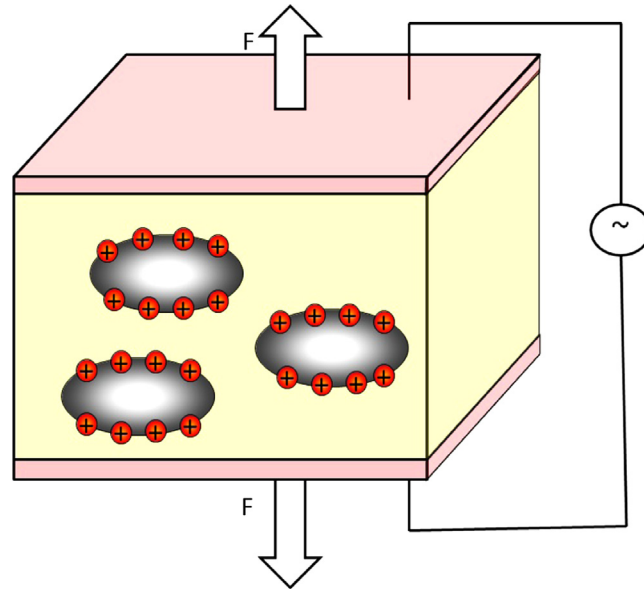
Fig. 3. An illustrative example of an electret material. Charge or dipoles are embedded in a homogeneous material. The inhomogeneity may be simply be due to voids and then usually charge is deposited on the void surfaces through a corona discharging process. Due to the Maxwell stress effect, an applied stress leads to a change in the pre-existing polarization thus causing the appearance of a linearized piezoelectric type coupling.

over the years (e.g. Dorfmann and Ogden, 2005; McMeeking and Landis, 2005; Suo et al., 2008; Steigmann, 2009; Eringen and Maugin, 1990; Liu, 2013a). In particular, in a series of works, Suo, Hong, Zhao and co-workers have explored a variety of topics related to soft electromechanical materials: instability, energy generation, theoretical formulations, etc. (Zhao and Suo, 2009; Koh et al., 2009; Wang et al., 2012).

We highlight here a concept related to the so-called electret materials which we will have occasion to analyze later in the paper. This concept illustrates an interesting use of the "Maxwell stress effect" to create soft "apparently piezoelectric" materials even though the actual materials are not intrinsically so. The idea is simple yet effective. Take a non-piezoelectric polymer. Embed layers of charges or dipoles in it. The embedded charge/dipole state is called the electret state. Assume that this state is reasonably stable (recall that charges will try to migrate to the surface and neutralize). Such a material behaves like a piezoelectric material provided the elastic properties in the material are non-uniform—e.g. multilayers with charge layers at the interfaces, or a foamy material containing charges on the void surfaces (Fig. 3). Due to the nonlinear Maxwell stress effect, deformation of the sample, induces a change in its pre-existing macroscopic polarization and similarly, a converse effect also exists. For all practical purposes, the "electret composite" behaves like a piezoelectric material—experimentally, a two-way linear coupling between stress-electric field can be detected even though microscopically the cause is the Maxwell stress effect. As will become evident through our mathematical derivations, a uniform homogeneous material with electrets will not exhibit this effect—inhomogeneity in material properties is a must. Usually, especially in cellular polymers like polypropylene, the air in the voids within the material is broken down through corona charging process thus depositing charge. Such prepared electret materials are found to be surprisingly stable for long durations of time. Experimental work has shown that electret-polypropylene foams can produce apparent piezoelectric coefficients up to 1200 pC/N—which is more than 6 times that of PZT (Hillenbrand and Sessler, 2008; Bauer et al., 2004)! Such "apparently piezoelectric" materials are especially desirable since they are highly flexible and are expected to have wide applications (Buchberger et al., 2008; Dansachmuller et al., 2007; Graz et al., 2006). To date, the work on this topic is primarily experimental in nature with only very simple models purporting to describe the experiments. In addition, the interaction of flexoelectricity (which leads to nanoscale size-effects) and electrets has remained unexplored.

In this paper we develop a nonlinear theoretical framework for flexoelectricity in soft materials—presented in Section 2. For transparency (without almost any loss of generality) we restrict ourselves to one-dimensional notation for most of the manuscript. In Section 3, we present a nonlinear derivation of the electret response and compare with some of the simpler approximate linear results. Apparently, a fully nonlinear solution to this problem has been conspicuously absent from the literature—somewhat surprising given that the targeted material systems undergo large deformations. We extend our earlier ideas of "creating apparently piezoelectric materials without using materials" to soft materials and illustrate this for a bilayer (Section 4) and subsequently explore the coupling between the Maxwell stress effect and flexoelectricity (Section 5). In particular, we illustrate an interesting nonlinear interplay between these two effects and electret states and show that unprecedentedly high values of electromechanical coupling can be achieved even in simple composite structures. Finally, predicated on a simple linearized theory of flexoelectricity in biological membranes, we present a few illustrative examples on its role in the linkages between electrical and mechanical stimuli.
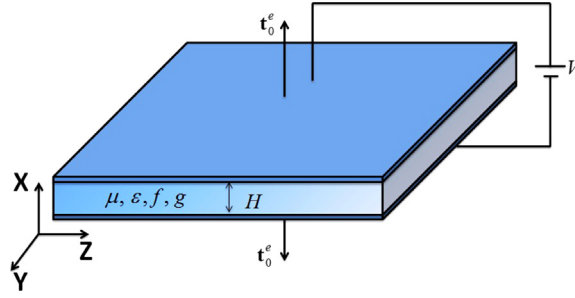
Fig. 4. The electro-elastic model: a film subject to an applied voltage.

2. Nonlinear formulation of flexoelectricity in soft materials

Consider a one-dimensional system (a thin film) shown in Fig. 4. Let (X, Y, Z) be the Lagrangian coordinates of material points, $P_0^e(X)$ be the external polarization (along X-direction, per unit volume), $\rho_0^e(X)$ be charge density (in the reference configuration), and $b_0^e(X)$, t_0^e be the body force, surface traction (equal and opposite at top and bottom faces) in X-direction, respectively. The electrodes on the top and bottom faces are assumed to be mechanically trivial. Nevertheless, the conducting electrodes maintain constant electrostatic potentials on both the top and bottom faces.

The electro-elastic state of the film is described by deformation and polarization—these are the independent variables in our formulation. In the presence of external charges, dipoles, applied voltage, and mechanical loads the film is deformed; the deformation is denoted by $(x, y, z) = \chi(X, Y, Z)$ with (x, y, z) being the Eulerian coordinates in the current configuration. Let P be the intrinsic polarization in X-direction per unit volume (in the reference configuration). Since the film is thin (the thickness H is much smaller than the width L in the other two directions) and transversely isotropic, for simplicity we restrict ourselves to the following class of deformation and polarization:

$$x = X + u(X), \quad y = Y\alpha(X), \quad z = Z\beta(X), \quad P = P(X), \tag{3}$$

where the scalar functions $u, \alpha, \beta, P : (0, H) \to \mathbb{R}$ are determined by the equilibrium conditions. We remark that the above kinematic assumption about the possible form of deformation is the equivalent to that in the classic theory of extension. Following the standard framework of nonlinear continuum mechanics, we introduce the *stretches* in X (resp. Y, Z)-direction: $\lambda_1 = 1 + \partial u / \partial X$ (resp. $\lambda_2 = \partial y / \partial Y = \alpha, \lambda_3 = \partial z / \partial Z = \beta$). Let $\mathbf{F} = \mathrm{Grad}\,\chi$ be the deformation gradient, $J = \det \mathbf{F} = \lambda_1 \lambda_2 \lambda_3$ be the Jacobian, and for brevity,

$$\lambda = (\lambda_1, \lambda_2, \lambda_3) \quad \text{and} \quad \mathbf{\Lambda} = (\Lambda_1, \Lambda_2, \Lambda_3) = \left(\frac{d\lambda_1}{dX}, \frac{d\lambda_2}{dX}, \frac{d\lambda_2}{dX} \right).$$

To model flexoelectricity we postulate that the internal/stored energy of the film is given by

$$U[\chi, P] = L^2 \int_0^H W(X; \lambda, \mathbf{\Lambda}, P)\, dX,$$

where the explicit X-dependence of W reflects that the film may be heterogeneous. Further, the polarized and deformed film induces electric field and interacts with external electrical and mechanical loading devices. To account for energies associated with electric fields and loading devices, we shall first solve for the electric field by the Maxwell equations, i.e., the electric field $e = -\xi_{,x}$ in the *current configuration* is determined by

$$\begin{cases} [-\varepsilon_0 \xi_{,x} + p(x) + p^e(x)]_{,x} = \rho^e(x) & \text{on } (0, h), \\ \xi(0) = 0, \quad \xi(h) = V, \end{cases} \tag{4}$$

where, without loss of generality, we have assumed that $x(0) = 0$, $x(H) = h$, and the intrinsic polarization (per unit volume), external polarization (per unit volume) and charge density in the *current configuration* are given by

$$p = \frac{P}{J}, \quad (\rho^e, p^e) = \frac{1}{J}(\rho_0^e, P_0^e). \tag{5}$$

Upon a change of variables, in the *reference configuration* we can rewrite the Maxwell equation (4) as

$$\begin{cases} \left[-\varepsilon_0 \frac{1}{\lambda_1} \xi_{,X} + \frac{P(X) + P_0^e(X)}{J} \right]_{,X} = \frac{\lambda_1}{J} \rho_0^e(X) & \text{on } (0, H), \\ \xi(0) = 0, \quad \xi(H) = V. \end{cases} \tag{6}$$

Taking into account of the energy of the electric field and external electrical and mechanical devices, we identify the *total* free energy of the system as (Liu, 2013a)

$$F[\chi, P] = U[\chi, P] + \mathcal{E}^{\text{elect}}[\chi, P] - L^2 \int_0^H b_0^e x(X)\, dX - L^2 [t_0^e(X) x(X)] \Big|_{X=0}^{X=H}, \tag{7}$$

where

$$\mathcal{E}^{\text{elect}}[\chi, P] = \frac{\varepsilon_0}{2} L^2 \int_0^h \frac{J}{\lambda_1} (\xi_{,x})^2\, dx + L^2 \left[\frac{J}{\lambda_1} \xi \left(-\varepsilon_0 \xi_{,x} + p + p^e \right) \right] \Big|_{x=0}^{x=h}$$

$$= \frac{\varepsilon_0}{2} L^2 \int_0^H \frac{J}{\lambda_1^2} (\xi_{,X})^2\, dX + L^2 \left[\frac{1}{\lambda_1} \xi \left(-\varepsilon_0 \frac{J}{\lambda_1} \xi_{,X} + P + P_0^e \right) \right] \Big|_{X=0}^{X=H}. \tag{8}$$

We remark that $\mathcal{E}^{\text{elect}}[\chi, P]$ is the electric energy, i.e., the sum of the electric field energy (the fringe field is neglected) and potential energy associated with the battery maintaining the potential difference, and that the second equality in (8) follows from a change of variable $x \to X$ and (5). Also, one recognizes the last two terms in (7) are the familiar potential energies contributed by the applied body force and surface tractions, respectively.

By the principle of minimum free energy, the equilibrium state of the film is determined by

$$\min_{(\chi, P) \in \mathcal{S}} F[\chi, P], \tag{9}$$

where the admissible space $\mathcal{S}$ of state variables include all deformations and polarizations (χ, P) satisfying (3).[3]

To find the Euler–Lagrange equations associated with a minimizer of (9), by (3) we consider variations

$$P \to P_\delta = P + \delta P_1, \quad \chi \to \chi_\delta = \begin{cases} X + u(X) + \delta u_1(X), \\ Y\alpha(X) + Y\delta\alpha_1(X), \\ Y\beta(X) + Z\delta\beta_1(X), \end{cases} \tag{10}$$

for arbitrary scalar functions $P_1, u_1, \alpha_1, \beta_1 : (0, H) \to \mathbb{R}$. Then standard variational calculations yield

$$\frac{1}{L^2} \frac{d}{d\delta} U[\chi_\delta, P_\delta] \Big|_{\delta=0} = \int_0^H \left[\frac{\partial W}{\partial \lambda} \cdot (u_{1,X}, \alpha_1, \beta_1) + \frac{\partial W}{\partial \Lambda} \cdot (u_{1,XX}, \alpha_{1,X}, \beta_{1,X}) + \frac{\partial W}{\partial P} P_1 \right]$$

$$= \int_0^H \left[-u_1 \tilde{T}_{1,X} + \alpha_1 \tilde{T}_2 + \beta_1 \tilde{T}_3 + \frac{\partial W}{\partial P} P_1 \right] + \left[\tilde{T}_1 u_1 + \frac{\partial W}{\partial \Lambda_2} \alpha_1 + \frac{\partial W}{\partial \Lambda_3} \beta_1 \right] \Big|_{X=0}^{X=H} + \frac{\partial W}{\partial \Lambda_1} u_{1,X} \Big|_{X=0}^{X=H} \tag{11}$$

where the second equality follows by integration by parts, and

$$\tilde{T}_i = \frac{\partial W}{\partial \lambda_i} - \frac{d}{dX} \frac{\partial W}{\partial \Lambda_i} \quad (i = 1, 2, 3) \tag{12}$$

is the "generalized" (normal) stress in a strain gradient theory. Further, to calculate the first variation of the electric energy $\mathcal{E}^{\text{elect}}(\chi_\delta, P_\delta)$, we write the associated electrostatic potential as

$$\xi_\delta = \xi + \delta \xi_1 + o(\delta). \tag{13}$$

From (4) and (8), and the detailed calculations in the Appendix, we find that

$$\mathcal{E}^{\text{elect}}(\chi_\delta, P_\delta) = \mathcal{E}^{\text{elect}}(\chi, P) + \delta \text{Var1} + o(\delta), \tag{14}$$

where

$$\text{Var1} = L^2 \int_0^H \left[P_1 \frac{\xi_{,X}}{\lambda_1} + u_{1,X} \tilde{\Sigma}_1 + \alpha_1 \tilde{\Sigma}_2 + \beta_1 \tilde{\Sigma}_3 \right] dX, \tag{15}$$

and

$$\tilde{\Sigma}_1 = -\frac{J}{\lambda_1^2} \xi_{,X} \left(-\frac{\varepsilon_0}{\lambda_1} \xi_{,X} + \frac{P + P_0^e}{J} \right) - \frac{\varepsilon_0 J}{2\lambda_1^3} (\xi_{,X})^2 = -\frac{1}{\lambda_1^2} \xi_{,X} (P + P_0^e) + \frac{\varepsilon_0 J}{2\lambda_1^3} (\xi_{,X})^2, \quad \tilde{\Sigma}_2 = -\frac{\varepsilon_0 J}{2\lambda_1^2 \lambda_2} (\xi_{,X})^2, \quad \tilde{\Sigma}_3 = -\frac{\varepsilon_0 J}{2\lambda_1^2 \lambda_3} (\xi_{,X})^2. \tag{16}$$

We remark that the above expression is the analogue of Piola stress of the familiar Maxwell stress which may be referred to as the *Piola–Maxwell* stress. A more systematic derivation of the *Piola–Maxwell* stress can be found in Tian (2007) and Liu (2013a).

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[3] The differentiability and integrability conditions are omitted for simplicity.

Collecting all terms contributed by the right-hand side of (7), by (11) and (15) we find the first variation of the total free energy is given by

$$\frac{1}{L^2}\frac{d}{d\delta}F[\chi_\delta, P_\delta]\Big|_{\delta=0} = \int_0^H \left[-u_1[(\tilde{T}_1+\tilde{\Sigma}_1)_{,X}+b_0^e]+\alpha_1(\tilde{T}_2+\tilde{\Sigma}_2)+\beta_1(\tilde{T}_3+\tilde{\Sigma}_3)+\left(\frac{\partial W}{\partial P}+\frac{\xi_{,X}}{\lambda_1}\right)P_1 \right]$$
$$+\left[\left(\tilde{T}_1+\tilde{\Sigma}_1-t_0^e\right)u_1\right]\Big|_{X=0}^{X=H} +\left[\frac{\partial W}{\partial \Lambda_2}\alpha_1+\frac{\partial W}{\partial \Lambda_3}\beta_1+\frac{\partial W}{\partial \Lambda_1}u_{1,X}\right]\Big|_{X=0}^{X=H}. \tag{17}$$

The equilibrium state (χ, P), i.e., a minimizer of (9), shall be such that the first variation (17) vanishes for arbitrary scalar functions $P_1, u_1, \alpha_1, \beta_1$, and henceforth necessarily satisfies the following system of ordinary differential equations:

$$\begin{cases} \dfrac{\partial W}{\partial P}+\dfrac{1}{\lambda_1}\xi_{,X}=0 & \text{on } (0,H), \\[2mm] [\tilde{T}_1+\tilde{\Sigma}_1]_{,X}+b_0^e=0 & \text{on } (0,H), \\[2mm] \tilde{T}_2+\tilde{\Sigma}_2=0 & \text{on } (0,H), \\[2mm] \tilde{T}_3+\tilde{\Sigma}_3=0 & \text{on } (0,H), \end{cases} \tag{18}$$

and the boundary conditions:

$$\begin{cases} [\tilde{T}_1+\tilde{\Sigma}_1-t_0^e]\Big|_{X=0\,\&\,H}=0, \\[2mm] \dfrac{\partial W}{\partial \Lambda_1}\Big|_{X=0\,\&\,H}=\dfrac{\partial W}{\partial \Lambda_2}\Big|_{X=0\,\&\,H}=\dfrac{\partial W}{\partial \Lambda_3}\Big|_{X=0\,\&\,H}=0. \end{cases} \tag{19}$$

We remark that (18) and (19), together with the Maxwell equation (6), form a closed differential system with five differential equations and five unknown scalar functions (i.e., ξ, P, u, α, β). In principle we can solve these equations for any physical quantities of interest. Also, this one dimensional theory is the electro-elastic counterpart of the classic nonlinear extension theory of a bar.

2.1. Isotropic nonlinear flexoelectric materials

To demonstrate the flexoelectric effects in soft matters, consider isotropic flexoelectric materials with the internal energy density given by

$$W(\lambda, \Lambda, P) = W_{\text{elast}}(\lambda) + \frac{g}{2}|\Lambda_1|^2 + f\Lambda_1 P + \frac{1}{2(\varepsilon-\varepsilon_0)J}|P|^2, \tag{20}$$

where $W_{\text{elast}}(\lambda)$ is the familiar strain energy density function dictating the mechanical properties of materials, the last term implies that the dielectric constant (i.e., permittivity) ε of the material is independent of deformation in the absence of flexoelectric effects, i.e., the third term $f\Lambda_1 P$, the second term $(g/2)|\Lambda_1|^2$ guarantees that the natural state $(\lambda, \Lambda, P)=(0,0,0)$ is the stable equilibrium state of the film in the absence of all external electrical and mechanical loads. Here, constants $f, g > 0$, $\varepsilon > \varepsilon_0$ are material properties which can be determined by benchmark experiments.

Inserting (20) into (12), we find that the generalized normal stress is given by

$$\tilde{T}_i = \frac{\partial W_{\text{elast}}}{\partial \lambda_i} - \delta_{1i}(g\Lambda_1 + fP)_{,X}.$$

Inserting (20) into (18)–(19) we obtain

$$\begin{cases} \dfrac{P}{(\varepsilon-\varepsilon_0)J}+f\Lambda_1+\dfrac{1}{\lambda_1}\dfrac{d\xi}{dX}=0 & \text{on } (0,H), \\[2mm] \left[\dfrac{\partial W_{\text{elast}}}{\partial \lambda_1}+\tilde{\Sigma}_1'-(gu_{,XX}+fP)_{,X}\right]_{,X}+b_0^e=0 & \text{on } (0,H), \\[2mm] \dfrac{\partial W_{\text{elast}}}{\partial \lambda_2}+\tilde{\Sigma}_2'=0 & \text{on } (0,H), \\[2mm] \dfrac{\partial W_{\text{elast}}}{\partial \lambda_3}+\tilde{\Sigma}_3'=0 & \text{on } (0,H), \end{cases} \tag{21}$$

and the boundary conditions:

$$\begin{cases} \left[\dfrac{\partial W_{\text{elast}}}{\partial \lambda_1}+\tilde{\Sigma}_1'-(gu_{,XX}+fP)_{,X}-t_0^e\right]\Big|_{X=0\,\&\,H}=0, \\[2mm] (gu_{,XX}+fP)\Big|_{X=0\,\&\,H}=0, \end{cases} \tag{22}$$

where

$$\tilde{\Sigma}_i^{'} = \tilde{\Sigma}_i - \frac{P^2}{2(\varepsilon - \varepsilon_0)J\lambda_i}.$$

We remark that if the elastic properties of the soft materials are specified, e.g. the Neo-Hookean hyperelastic model with (μ —shear modulus, κ—bulk modulus)

$$W_{\text{elast}}(\lambda) = \frac{\mu}{2}\left[J^{-2/3}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2) - 3\right] + \frac{\kappa}{2}(J - 1)^2, \tag{23}$$

then Eqs. (21)–(22) together with (6) form a closed boundary value problem which can be solved to determine the state variables (χ, P) (i.e., u, α, β, P) and electrostatic potential ξ.

Moreover, if the material is *incompressible* with $J = \lambda_1 \lambda_2 \lambda_3 = 1$, the state variables as given by (3) shall satisfy

$$J = (1 + u_X)\alpha\beta = 1.$$

To explore the implication of the above constraint, by the method of Lagrangian multiplier we add to the total free energy (7) a term

$$\int_0^H q[(1 + u_X)\alpha\beta - 1] \, dX,$$

where $q : (0, H) \to \mathbb{R}$ can be interpreted as the hydrostatic pressure as in the classic context. Repeating the calculations from (10) to (17) and taking into account the contribution of the above additional term, we find the associated Euler–Lagrange equations

$$\begin{cases} \dfrac{\partial W}{\partial P} + \dfrac{1}{\lambda_1}\xi_{,X} = 0 & \text{on } (0, H), \\[2mm] \left[\tilde{T}_1 + \tilde{\Sigma}_1 + \dfrac{q}{\lambda_1}\right]_{,X} + b_0^e = 0 & \text{on } (0, H), \\[2mm] \tilde{T}_2 + \tilde{\Sigma}_2 + \dfrac{q}{\lambda_2} = 0 & \text{on } (0, H), \\[2mm] \tilde{T}_3 + \tilde{\Sigma}_3 + \dfrac{q}{\lambda_3} = 0 & \text{on } (0, H), \end{cases} \tag{24}$$

and the boundary conditions:

$$\begin{cases} \left[\tilde{T}_1 + \tilde{\Sigma}_1 + \dfrac{q}{\lambda_1} - t_0^e\right]\Big|_{X = 0 \text{ & } H} = 0, \\[2mm] \dfrac{\partial W}{\partial \Lambda_1}\Big|_{X = 0 \text{ & } H} = \dfrac{\partial W}{\partial \Lambda_2}\Big|_{X = 0 \text{ & } H} = \dfrac{\partial W}{\partial \Lambda_3}\Big|_{X = 0 \text{ & } H} = 0. \end{cases} \tag{25}$$

For incompressible flexoelectric Neo-Hookean material described by (20) and (23), based on symmetry we observe that a solution of the above boundary value problem (24) and (25) and the Maxwell equation (6) shall satisfy

$$\lambda_2 = \alpha = \lambda_3 = \beta = \lambda_1^{-1/2} \quad \text{on } (0, H). \tag{26}$$

Further, we can rewrite the strain energy density (23) as

$$W_{\text{elast}}(\lambda_1) = \frac{\mu}{2}\left(\lambda_1^2 + \frac{2}{\lambda_1} - 3\right), \tag{27}$$

which, by (12), $(24)_{3,4}$ and (26), implies

$$\tilde{T}_2 = \tilde{T}_3 = 0, \quad \tilde{\Sigma}_2 = \tilde{\Sigma}_3 = -q\lambda_1^{1/2} \quad \text{on } (0, H).$$

Eliminating q in $(24)_{1,2}$ by the above equation, we obtain the following explicit boundary value problem for u, ξ, P:

$$\begin{cases} \dfrac{P}{(\varepsilon - \varepsilon_0)} + f\lambda_{1,X} + \lambda_1^{-1}\xi_{,X} = 0 & \text{on } (0, H), \\[2mm] [\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq} - (gu_{,XX} + fP)_{,X}]_{,X} + b_0^e = 0 & \text{on } (0, H), \\[2mm] [-\varepsilon_0\lambda_1^{-1}\xi_{,X} + P + P_0^e]_{,X} = \lambda_1\rho_0^e & \text{on } (0, H), \\[2mm] \xi(0) = 0, \quad \xi(H) = V, \quad (gu_{,XX} + fP)|_{X = 0 \text{ & } H} = 0, \\[2mm] [\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq} - (gu_{,XX} + fP)_{,X} - t_0^e]|_{X = 0, \text{ & } H} = 0, \end{cases} \tag{28}$$

where $\lambda_1 = 1 + u_X$,

$$\tilde{\Sigma}_{eq} = \tilde{\Sigma}_1 - \lambda_1^{-3/2}\tilde{\Sigma}_2 = -\frac{1}{\lambda_1^2}\xi_{,X}(P + P_0^e) + \frac{\varepsilon_0}{\lambda_1^3}(\xi_{,X})^2.$$

In the absence of flexoelectricity ($f = g = 0$), the above boundary value problem can be rewritten as

$$
\begin{cases}
\dfrac{P}{(\varepsilon - \varepsilon_0)} + \lambda_1^{-1} \xi_X = 0 & \text{on } (0, H), \\[2mm]
[\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq}]_X + b_0^e = 0 & \text{on } (0, H), \\[2mm]
[-\varepsilon_0 \lambda_1^{-1} \xi_X + P + P_0^e]_X = \lambda_1 \rho_0^e & \text{on } (0, H), \\[2mm]
\xi(0) = 0, \quad \xi(H) = V, \\[2mm]
[\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq} - t_0^e]|_{X = 0 \,\&\, H} = 0.
\end{cases}
\tag{29}
$$

In the absence of external dipoles, charges and body force, i.e., $P_0^e = \rho_0^e = b_0^e = 0$, upon a change of variable $X \to x$ the above equation can be rewritten as

$$
\begin{cases}
\dfrac{p}{(\varepsilon - \varepsilon_0)} + \xi_{,x} = 0 & \text{on } (0, h), \\[2mm]
[\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq}]_X = 0 & \text{on } (0, h), \\[2mm]
[-\varepsilon_0 \xi_{,x}]_x = 0 & \text{on } (0, h), \\[2mm]
\xi(0) = 0, \quad \xi(h) = V, \\[2mm]
[\mu(\lambda_1 - \lambda_1^{-2}) + \tilde{\Sigma}_{eq} - t_0^e]|_{X = 0 \,\&\, H} = 0.
\end{cases}
\tag{30}
$$

A solution to the above problem is obviously given by $\xi(x) = (x/h)V$, and

$$
\mu(\lambda_1 - \lambda_1^{-2}) = -\tilde{\Sigma}_{eq} = t_0^e - \frac{1}{\lambda_1} \varepsilon \left(\frac{V}{h}\right)^2 = t_0^e - \frac{1}{\lambda_1^3} \varepsilon \left(\frac{V}{H}\right)^2.
\tag{31}
$$

We comment that the term $-\varepsilon(V/h)^2$ is often termed as "the equivalent electromechanical pressure" in the engineering literature. For small strain ($(h - H)/H \ll 1$ (which requires $t_0^e \ll \mu$), we immediately have

$$
\lambda_1 - 1 = \frac{h - H}{H} \approx \frac{1}{3\mu} \left[t_0^e - \varepsilon \left(\frac{V}{H}\right)^2 \right].
\tag{32}
$$

3. Nonlinear behavior of electrets: creating soft piezoelectric materials

In this section, we study an electret composite as discussed in the Introduction. We chose the simplest possible configuration: a simple bilayer with charge embedded at the interface. Using crude approximations, this problem has been earlier studied by Kacprzyk et al. (1995)—the analytical models are fairly simple and the role of Maxwell stress is not evident. A discussion of the experimental work may be found in Hillenbrand and Sessler (2008) and Bauer et al. (2004). As shown in Fig. 5, let $X = H_a$ and $X = -H_b$ correspond to the top and bottom surfaces, respectively. The interface between two layers locates at $X = 0$. Then the charge density can be written as $\rho_0^e(X) = q_0 \delta(X)$. $\delta(X)$ is the Dirac delta function which is only non-zero at $X = 0$.

Below we present a nonlinear solution of this problem by the theory developed in the last section—in this section, we ignore flexoelectricity. Without loss of generality, assume that the deformation $x : (-H_b, H_a) \to \mathbb{R}$ and electrostatic potential $\xi : (-H_b, H_a) \to \mathbb{R}$ satisfy

$$
x(X = 0), \quad x(X = H_a) = h_a, \quad x(X = -H_b) = -h_b,
$$

$$
\xi(X = 0) = 0, \quad \xi(X = H_a) = V_a, \quad \xi(X = -H_b) = -V_b,
$$

where $h_i, V_i \in \mathbb{R}$ ($i = a, b$) are constants to be determined. By $(29)_{1,3,4}$ we have

$$
V_a + V_b = V, \quad \frac{\varepsilon_a}{\lambda_a^2} \frac{V_a}{H_a} - \frac{\varepsilon_b}{\lambda_b^2} \frac{V_b}{H_b} = q_0
\tag{33}
$$

In the absence of flexoelectricity ($f = g = 0$), by (30)–(31) we have[4]

$$
\mu_i(\lambda_i - \lambda_i^{-2}) = t_0^e - \frac{1}{\lambda_i^3} \varepsilon_i \left(\frac{V_i}{H_i}\right)^2 \quad (i = a, b).
\tag{34}
$$

We remark that (33) and (34) are nonlinear algebraic equations which can be conveniently solved numerically.

Although we will eventually present nonlinear numerical results, it is instructive to obtain an approximate analytical solution also; which we do through linearization. For that purpose, with the assumption of small strains, by (32) equations

---

[4] Subsequently we drop the subscript '1' associated with stretching λ_1 in X-direction; the subscript 'i' now represents the phase a or b.
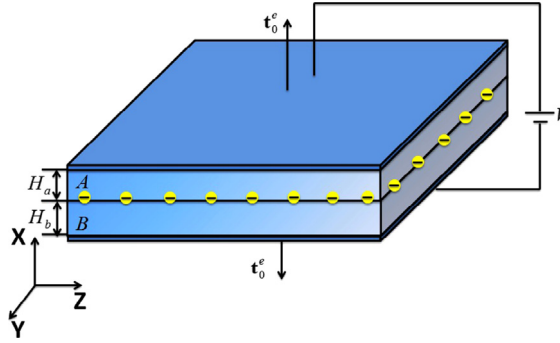
Fig. 5. The electret composite: a bilayer with embedded charge at the interface.

in (34) imply:

$$\lambda_i - 1 = \frac{h_i - H_i}{H_i} \approx \frac{1}{3\mu_i}\left[t_0^e - \varepsilon_i\left(\frac{V_i}{H_i}\right)^2\right]. \tag{35}$$

Solving (33) and (35) for V_j, h_j with the assumption of small strains ($\lambda_a, \lambda_b \to 1$), we obtain

$$V_a = H_a\frac{q_0 H_b + V\varepsilon_b}{\varepsilon_a H_b + \varepsilon_b H_a}, \quad V_b = H_b\frac{-q_0 H_a + V\varepsilon_a}{\varepsilon_a H_b + \varepsilon_b H_a}, \quad h_a = H_a + \frac{H_a}{3\mu_a}\left(-\varepsilon_a\left(\frac{V_a}{H_a}\right)^2\right), \quad h_b = H_b + \frac{H_b}{3\mu_b}\left(-\varepsilon_b\left(\frac{V_b}{H_b}\right)^2\right). \tag{36}$$

Therefore, the total change of the thickness Δu is given by

$$\Delta u = h_a + h_b - H_a - H_b = -\frac{1}{3\mu_a}\varepsilon_a\frac{q_0^2 H_b^2 H_a}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{1}{3\mu_b}\varepsilon_b\frac{q_0^2 H_a^2 H_b}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{2}{3\mu_a}\varepsilon_a\frac{q_0 H_a H_b \varepsilon_b V}{(\varepsilon_a H_b + \varepsilon_b H_a)^2}$$

$$+ \frac{2}{3\mu_b}\varepsilon_b\frac{q_0 H_a H_b \varepsilon_a V}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{1}{3\mu_a}\varepsilon_a\frac{V^2\varepsilon_b^2 H_a}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{1}{3\mu_b}\varepsilon_b\frac{V^2\varepsilon_a^2 H_b}{(\varepsilon_a H_b + \varepsilon_b H_a)^2}.$$

For physical transparency, it is convenient to separate Δu into two parts:

$$\Delta u = \Delta u^{em} + \Delta u^m,$$

where Δu^{em} is the displacement caused by the interaction of the electret state and Maxwell stress and is linearly proportional to the applied voltage, and Δu^m is the displacement proportional to square of the applied voltage (similar to what would happen for a homogenous material)

$$\Delta u^{em} = -\frac{1}{3\mu_a}\varepsilon_a\frac{q_0^2 H_b^2 H_a}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{1}{3\mu_b}\varepsilon_b\frac{q_0^2 H_a^2 H_b}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{2}{3\mu_a}\varepsilon_a\frac{q_0 H_a H_b \varepsilon_b V}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} + \frac{2}{3\mu_b}\varepsilon_b\frac{q_0 H_a H_b \varepsilon_a V}{(\varepsilon_a H_b + \varepsilon_b H_a)^2},$$

$$\Delta u^m = -\frac{1}{3\mu_a}\varepsilon_a\frac{V^2\varepsilon_b^2 H_a}{(\varepsilon_a H_b + \varepsilon_b H_a)^2} - \frac{1}{3\mu_b}\varepsilon_b\frac{V^2\varepsilon_a^2 H_b}{(\varepsilon_a H_b + \varepsilon_b H_a)^2}.$$

Similar to the form reported in the literature (Kacprzyk et al., 1995), we then define the *linear* change of the thickness with respect to the applied voltage as the *effective piezoelectric coefficient*:

$$d^{\text{eff}} = \frac{d\Delta u^{em}}{dV} = -\frac{2q_0 H_a H_b \varepsilon_a \varepsilon_b}{3(\varepsilon_a H_b + \varepsilon_b H_a)^2}\left(\frac{1}{\mu_a} - \frac{1}{\mu_b}\right). \tag{37}$$

However, for soft material, the small-deformation assumption is certainly contraindicated. In the following, we present results for the full nonlinear problem (albeit using numerics). A general purpose finite-element based partial differential equation solver (COMSOL4.3a, 2012) was used for this purpose. Because of the highly nonlinear nature of the problem, the quartic (4th-order) 1D finite element is used for all the calculations in this paper. Another reason of using the higher order element here is that the high order continuity needs to be satisfied throughout the specimen. Polypropylene cellular film and polyvinylidene fluoride (PVDF) are used for layers A and B, respectively. The material properties listed below are from the literatures (Qu and Yu, 2011; Chu and Salem, 2012).

Layer A: $\mu_a = 0.95$ MPa, $\varepsilon_a = 2.35\varepsilon_0$, $\varepsilon_0 = 8.85 \times 10^{-12} F/m$.

Layer B: $\mu_b = 2$ GPa, $\varepsilon_b = 9.5\varepsilon_0$.

Here, μ_i and ε_i ($i = a, b$) are the shear modulus and permittivity of the A-layer and B-layer, respectively.

It is worthwhile to note that while in the linearized analytical solution, we are able to extract the "piezoelectric type" response by discarding the pure Maxwell stress effect (i.e., the quadratic term in the voltage), in the numerical solution we resort to the following: two cases for $q_0 = -1$ mC/m² and $q_0 = 0$ are calculated separately. The charge density value used for
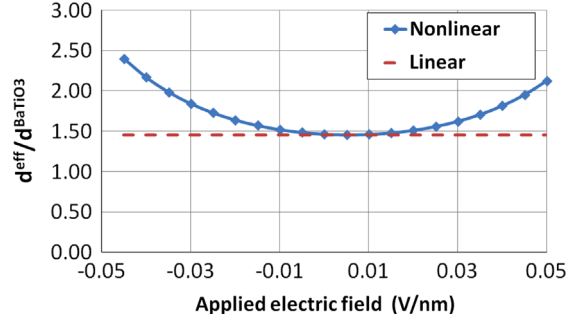
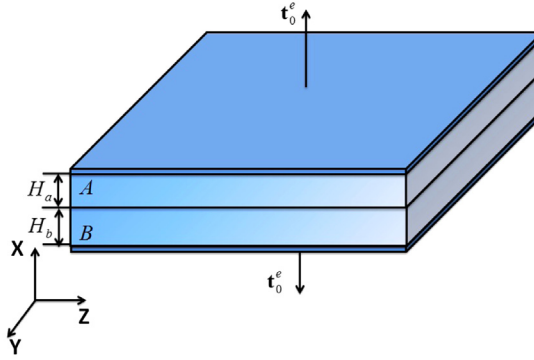Fig. 6. Effective piezoelectricity of a soft electret composite.

Fig. 7. A bilayer flexoelectric structure under uniform mechanical loading.

the numerical results is of the same order as what is realized and measured experimentally (Qu and Yu, 2011; Sessler and Hillenbrand, 1999). Then the ratio of the change of the total thickness Δu to the change of applied voltage ΔV is computed for each case. The effective piezoelectricity d^{eff} is then the difference between the $\Delta u / \Delta V$ from the two cases. Fig. 6 shows the variation of the effective piezoelectric coefficient with respect to the applied electric field. In the figure, d^{eff} is normalized by the piezoelectric coefficient of Barium Titanate d^{BaTiO_3} which is about 78 pC/N. When the applied electric field is small, the predicted d^{eff} is very close to the linear model. However, as evident, the linear model is valid for a very narrow range of applied electric field. A remarkable observation is that when the applied electric field is as high as 0.05 V/nm, we can obtain an effective piezoelectric response which is more than twice that of Barium Titanate. As evident from the linear model (37), by choosing a foam (i.e., polymer+voids rather than two polymers) we can engineer an even stronger response.

4. Nonlinear flexoelectricity: creating soft piezoelectric materials

In this section, we consider the flexoelectric effect in the context of the simple bilayer structure shown in Fig. 7. The difference between this model and the previous one is that external interfacial charge is absent. As explained in the Introduction, the two layers will experience different deformations and result in strain gradients at the interface. Flexoelectricity will then cause the bilayer structure to behave like an apparently piezoelectric material. For this problem, the voltage difference between the two surfaces is maintained to be zero throughout the calculation; however, a uniform loading t_0^e is applied. Under this loading, an electric displacement will ensue in the body. The effective piezoelectricity is then calculated by $d^{\text{eff}} = d\tilde{D} / dt_0^e$.

In the absence of external polarization, charges and body force, i.e., $P_0^e = b_0^e = 0$, by (28) we obtain the following explicit boundary value problem for u, ξ, P:

$$
\begin{cases}
\dfrac{P}{(\varepsilon - \varepsilon_0)} + f\lambda_{,X} + \lambda^{-1}\xi_{,X} = 0 & \text{on } (-H_b, H_a), \\[2mm]
[\mu(\lambda - \lambda^{-2}) + \tilde{\Sigma}_{eq} - (gu_{,XX} + fP)_{,X}]_{,X} = 0 & \text{on } (-H_b, H_a), \\[2mm]
[-\varepsilon_0 \lambda^{-2}\xi_{,X} + \lambda^{-1}P]_{,X} = 0 & \text{on } (-H_b, 0) \cup (0, H_a), \\[2mm]
\xi(-H_b) = 0, \quad \xi(H_a) = 0, \quad (gu_{,XX} + fP)|_{X = -H_b \ \& \ H_a} = 0, \\[2mm]
[\mu(\lambda - \lambda^{-2}) + \tilde{\Sigma}_{eq} - (gu_{,XXX} + fP_{,X}) - t_0^e]|_{X = -H_b \ \& \ H_a} = 0,
\end{cases}
\tag{38}
$$

where

$$\lambda = 1 + u_{,X}, \quad \tilde{\Sigma}_{eq} = -\frac{1}{\lambda^2}\xi_{,X}(P + P_0^e) + \frac{\varepsilon_0}{\lambda^3}(\xi_{,X})^2.$$

The interfacial conditions are

$$\begin{cases} [\![\xi]\!] = 0, \quad [\![-\varepsilon_0\lambda^{-2}\xi_{,X} + \lambda^{-1}P]\!] = 0, \\ [\![u]\!] = 0, \quad [\![u_{,X}]\!] = 0, \quad [\![gu_{,XX} + fP]\!] = 0, \\ [\![\mu(\lambda - \lambda^{-2}) + \tilde{\Sigma}_{eq} - (gu_{,XX} + fP)_{,X}]\!] = 0, \end{cases}$$

where $[\![\ ]\!] = ()|_{X=0+} - ()|_{X=0-}$.

For this problem, we set the displacement on the interface $u(0)$ to be zero to eliminate rigid body motion. Polypropylene cellular film and polyvinylidene fluoride (PVDF) are used for the layers A and B, respectively—as in the preceding section. The material properties of the two layers are given by:

Layer A: $\mu_a = 0.95$ MPa, $f_a = 46.79$ N m/C, $g_a = 1.28 \times 10^{-8}$ N, $\varepsilon_a = 2.35\varepsilon_0$.

Layer B: $\mu_b = 2$ GPa, $f_b = 179$ N m/C, $g_b = 5.42 \times 10^{-7}$ N, $\varepsilon_b = 9.5\varepsilon_0$.

The flexoelectric coefficient of PVDF is reported in Chu and Salem (2012). Since there is no report on the flexoelectricity coefficient of polypropylene cellular film, in this work, we have assumed a reasonable value which is within the range of known values for common polymer materials. Also, the value of g has to be estimated. The reader is referred to two works (Maranganti and Sharma, 2007; Nikolov et al., 2007) that use atomistic and microscopic considerations to determine this parameter that sets the nonlocal elastic length scale. We use a simpler route to predict an approximate value for the polymers studied by us. As motivated by Maranganti and Sharma (2007), the characteristic nonlocal elastic length scale can be approximated by the radius of gyration. Accordingly, we set $\sqrt{g/3\mu} \sim R_g$, where R_g is the radius of gyration of the polymers studied.

Fig. 8 shows the size effect on the effective piezoelectricity for different stress levels. In this figure, once again, d^{eff} is normalized by $d^{\mathrm{BaTiO_3}}$. Several observations may be made: (i) unlike the size-independent electret case studied in the preceding section, scaling can be profitably used to engineer a high electromechanical coupling by exploiting the flexoelectric effect, (ii) the resulting electromechanical coupling in soft materials is *quite high*—reaching close to 20 times that of Barium Titanate!, and finally, (iii) unlike hard ceramics, the pronounced size-effect is evident at the micron scale (as opposed to nanoscale). The latter has important ramifications in terms of experimental verification and exploitation for practical applications.

5. The interaction of electrets and flexoelectricity

From the preceding sections, it is well apparent that electrets and flexoelectricity both may be used to dramatically enhance electromechanical coupling in soft materials—in particular, this enhanced electromechanical coupling is not the restrictive one-way coupling characteristic of the Maxwell stress effect or electrostriction but rather something that manifests itself as macroscopic piezoelectricity. In this section, we consider the likelihood of a nonlinear interaction between these effects. The statement of this boundary value problem is almost the same as the previous section except that there is a jump of electric displacement at the interface—since we now embed an interfacial external charge between the layers:

$$[\![-\varepsilon_0\lambda^{-2}\xi_{,X} + \lambda^{-1}P]\!] = q_0$$

where q_0 is the charge density at the interface.

Calculations are done numerically as before. As shown in Fig. 9(a), flexoelectricity dominates completely at small sizes. Only at very large sizes (where the flexoelectric response asymptotically goes to zero) does the electret effect show any impact (Fig. 9(b)). It is found that when above 20 μm, the "no electret" model shows no piezoelectricity while the "electret" model shows a constant piezoelectricity which is size-independent. We conclude, from our results that while both these
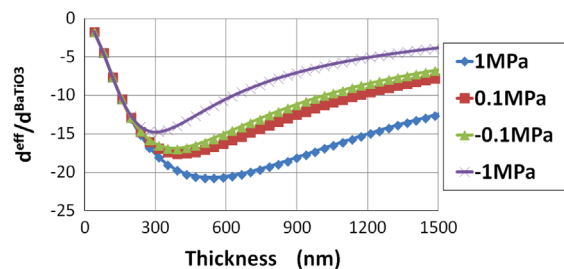

Fig. 8. Size-dependent effective piezoelectricity as a function of applied loading: t_0^e.
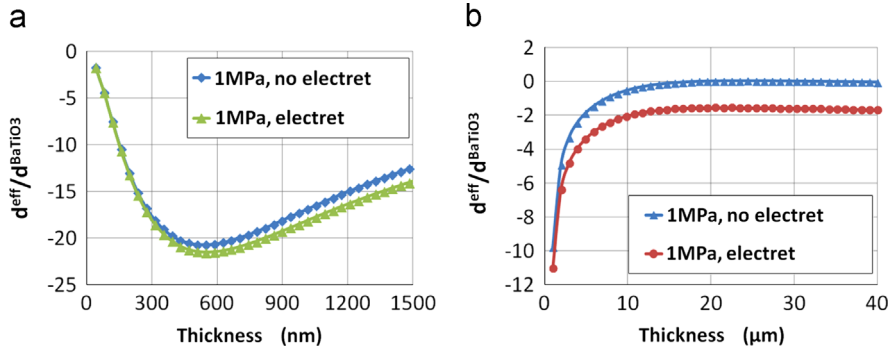
Fig. 9. Interaction between the flexoelectric and electret effects at (a) nanometer scale and (b) micrometer scale.

effects are important in their own right, there is little interaction between them (at least in the one-dimensional problems studied by us).

6. A simplified theory for biological membranes and illustrative examples

It is anticipated that the coupling between elasticity and electricity in biological membranes is important for many biological functions. From a mechanistic viewpoint, we can rule out the piezoelectric effect in biological membranes by symmetry. Therefore, the leading order coupling between strain and polarization has to be "flexoelectric", i.e., the coupling between strain gradient and polarization.

The flexoelectric theory concerning thin membranes has been developed in Mohammadi et al. (2014); a formal derivation of the theory is presented in Liu. In this formulation, we consider a thin membrane occupying $U \times (-h/2, h/2) \subset \mathbb{R}^3$, with $U \subset \mathbb{R}^2$ being an open bounded domain in xy-plane and h being the thickness of the membrane. Since the thickness $h \gg 1$, the thin membrane may be idealized as a two-dimensional body; the thermodynamic state is then described by the out-of-plane displacement $w : U \to \mathbb{R}$ and the out-of-plane polarization $P : U \to \mathbb{R}$. For thin membranes, we also anticipate that bending is the predominant mode of deformation, and hence use the linearized curvature tensor (or strain gradient) of the membrane $\xi = -\nabla\nabla w$ to describe the deformed state of the membrane.

To model the flexoelectric effect, we postulate that the total internal/stored energy of an isotropic membrane is given by

$$U[w, P] = \int_U W(\nabla\nabla w, P), \tag{39}$$

where $W : \mathbb{R}_{\text{sym}}^{2\times2} \times \mathbb{R} \to \mathbb{R}$ is the total internal energy density function and given by a quadratic function

$$W(\nabla\nabla w, P) = \frac{\kappa_b}{2}(\Delta w)^2 + \kappa_g \det(\nabla\nabla w) + fP\Delta w + \frac{1}{2}a_z P^2. \tag{40}$$

Here, we notice that the elastic part, $(\kappa_b/2)(\Delta w)^2 + \kappa_g \det(\nabla\nabla w)$, of the membrane energy coincides with the linearized Helfrich–Canham model (Helfrich, 1973) of biological membranes that are widely used by bio-physicists (which is in turn identical to the Kirchhoff-Love plate theory (Love, 1944)), the term $fP\Delta w$ gives rise to the coupling between polarization and curvature (flexoelectric effects), and the last term $\frac{1}{2}a_z P^2$ arises from the dielectric property of the membrane. The constants $\kappa_b, \kappa_g, f, a_z$ are material properties of the flexoelectric membrane and may in general depend on in-plane positions. Moreover, the stability of the membrane requires that (Mohammadi et al., 2014):

$$\kappa_b > 0, \quad -2\kappa_b < \kappa_g < 0, \quad a > 0 \quad \text{and} \quad \kappa_b + \frac{\kappa_g}{2} > \frac{f^2}{a}. \tag{41}$$

The boundary of the flexoelectric membrane is clamped: $w, \nabla w|_{\partial U} = 0$. Then under the application of an external electric field $E_z : U \to \mathbb{R}$ and a mechanical body force $b_z : U \to \mathbb{R}$, the total free energy of the membrane is given by

$$F[w, P] = \int_U W(\nabla\nabla w, P) - \int_U [PE_z + wb_z], \tag{42}$$

where the first integral is the internal energy of the flexoelectric membrane, and the second one is the potential energy arising from the interaction between the membrane and the external electric field and mechanical loading device.

In the equilibrium state, by the principle of minimum free energy the pair of (w, P) shall minimize the total free energy (42):

$$\min_{(w,P) \in \mathcal{S}} F[w, P] \tag{43}$$

where the admissible space for (w, P) is given by

$$\mathcal{S} := \left\{ (w, P) : \int_U |\nabla\nabla w|^2 < +\infty, \ \int_U |P|^2 < +\infty, \ w, \nabla w|_{\partial U} = 0 \right\}. \tag{44}$$

By standard variational calculations (Mohammadi et al., 2014), it can be shown that a minimizer (w, P) of the minimization problem (43) necessarily satisfies the following Euler–Lagrange equations and boundary conditions:

$$\begin{cases} \nabla\nabla \cdot (\mathbf{L}\nabla\nabla w) + \Delta(fP) - b_z = 0 & \text{on } U, \\ f\Delta w + aP - E_z = 0 & \text{on } U, \\ w = 0, \quad \nabla w = 0 & \text{on } \partial U. \end{cases} \tag{45}$$

Using $(45)_2$ we eliminate P in $(45)_{1, \ 4,5}$, and obtain

$$\begin{cases} \mathbf{M} := \tilde{\mathbf{L}}\nabla\nabla w, \quad \tilde{\mathbf{L}} := \mathbf{L} - \dfrac{f^2}{a}\mathbf{I} \otimes \mathbf{I} & \text{on } U, \\ \nabla\nabla \cdot \left(\mathbf{M} + \dfrac{f}{a}E_z\mathbf{I} \right) - b_z = 0 & \text{on } U, \\ w = 0, \quad \nabla w = 0 & \text{on } \partial U. \end{cases} \tag{46}$$

6.1. Bending of flexoelectric membranes in an electric field

Consider a flexoelectric membrane. In the absence of applied body force $b_z = 0$, the membrane can nevertheless be bent by an external electric field since the term $\gamma E_z\mathbf{I}$ in $(46)_1$ serves as a "source" term for the boundary value problem (46) of w. It is of interest to investigate the effects of the external field on w. For simplicity, assume that the membrane is homogeneous and isotropic, and hence the boundary value problem (46) can be rewritten as

$$\Delta[k_b\Delta w + \gamma E_z] = 0 \quad \text{on } U, \tag{47}$$

where $k_b = 2\mu_b + \lambda_b - f^2/a$. For appropriate boundary conditions as specified in $(40)_{3,4}$, it is standard to solve (47) for w. As examples, below we present a few explicit solutions, assuming infinite membrane on $\mathbb{R}^2$ and natural boundary conditions at the infinity:

$$|\nabla\nabla w(\mathbf{x})| \to 0 \quad \text{as } |\mathbf{x}| \to +\infty. \tag{48}$$

We remark that w is only determined within an arbitrary linear function of (x, y) by the above conditions. Also, appropriate decay conditions on the source term E_z are required for (48). These simple solutions may be used for measuring the material properties in (40) and as the benchmarks of numerical schemes.

6.2. $E_z = E_z(x)$

Since E_z is independent of y, by symmetry we seek a solution of form $w = w(x)$ to (47), i.e.,

$$\frac{d^2}{dx^2}\left[k_b\frac{d^2}{dx^2}w(x) + \gamma E_z(x) \right] = 0 \quad \forall x \in \mathbb{R}.$$

The general solution to the above equation is given by

$$w(x) = -\iint \frac{\gamma}{k_b}E_z(x) + C_0 + C_1 x, \tag{49}$$

where C_0, C_1 are the integration constants and shall be determined by boundary conditions. In particular, if the external field is generated by an infinite line charge of line density q along $\mathbf{e}_y$ direction and above the membrane with distance z_0, then

$$E_z(x) = -\frac{qz_0}{2\pi\varepsilon_0}(z_0^2 + x^2)^{-1},$$

where ε_0 is the permittivity of vacuum. By (49) we have

$$w(x) = \frac{q\gamma}{2\pi\varepsilon_0 k_b}\left[x\arctan\left(\frac{x}{z_0}\right) - \frac{z_0}{2}\log(x^2 + z_0^2) \right].$$

Fig. 10 shows that the line charge q can cause the deformation of a flexoelectric membrane. In the figure, x is normalized by the distance between the charge and the membrane z_0, $w(x)$ is normalized by the term $w_0 = q\gamma/\varepsilon_0 k_b z_0$. We expect this and related results to have important implications for the study of lipid bilayers by ions in the surrounding electrolytes.
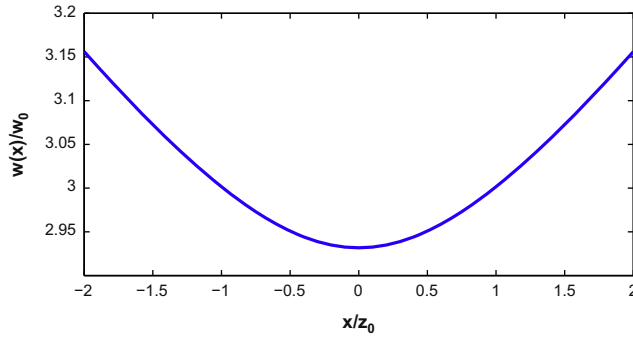
Fig. 10. Deflection of a flexoelectric membrane near a line charge.

6.3. $E_z = E_z(r)$

Since E_z is only a function of $r = (x^2 + y^2)^{1/2}$, by symmetry we seek a solution $w = w(r)$ to (47):

$$\frac{d}{r\,dr} r \frac{d}{dr} \left[k_b \frac{d}{r\,dr} r \frac{d}{dr} w(r) + \gamma E_z(r) \right] = 0 \quad \forall r > 0.$$

Neglecting immaterial integration constants, by (48) we have

$$\frac{d}{r\,dr} r \frac{d}{dr} w(r) = -\frac{\gamma}{k_b} E_z(r). \tag{50}$$

Upon specifying the functional form $E_z(r)$, we can integrate the above equation explicitly for $w(r)$. In particular, if the external field is generated by a point charge Q above the membrane at $z = z_0$, then

$$E_z(r) = -\frac{qz_0}{4\pi\varepsilon_0} (z_0^2 + r^2)^{-3/2}.$$

By (50) we have

$$w(r) = -\frac{q\gamma}{4\pi\varepsilon_0 k_b} \left[\log(r) - \log\left(z_0^2 + z_0 \sqrt{r^2 + z_0^2} \right) \right].$$

6.4. $E_z = \overline{\mathbf{E}}_z \chi_{\Omega_1}$

Here $\Omega_1 \subset \mathbb{R}^2$ is a regular domain; χ_{Ω_1}, equal to one on Ω_1 and zero otherwise, is the characteristic function of Ω_1. By (48), a solution to (46) necessarily satisfies

$$\Delta w = -\frac{\gamma}{k_b} \overline{\mathbf{E}}_z \chi_{\Omega_1} \quad \text{on } \mathbb{R}^2. \tag{51}$$

If $\overline{\mathbf{E}}_z$ is constant and Ω_1 is an ellipse, it is well-known that a solution to the above equation is necessarily a quadratic function restricted to Ω_1 (Kellogg, 1954). In other words,

$$\nabla\nabla w = -\frac{\gamma}{k_b} \overline{\mathbf{E}}_z \mathbf{Q} \quad \text{on } \Omega_1, \tag{52}$$

where the matrix $\mathbf{Q} \in \mathbb{R}^{2\times2}_{\text{sym}}$ is given by elliptic integrals (Liu et al., 2006; Liu, 2013b):

$$\mathbf{Q} = \frac{1}{2\pi} \int_{S^1} \frac{\hat{\mathbf{k}} \otimes \hat{\mathbf{k}} \det(\mathbf{A})}{|\mathbf{A}\hat{\mathbf{k}}|^2} d\hat{\mathbf{k}},$$

where $\mathbf{A} \in \mathbb{R}^{2\times2}_{\text{sym}}$ is such that $\Omega_1 = \mathbf{Ax} : \mathbf{x} \in B_2$, $B_2 \subset \mathbb{R}^2$ is a unit circle, and $S^1 = \mathbf{x} \in \mathbb{R}^2 : |\mathbf{x}| = 1$ is the perimeter of the unit circle.

Physically, Eq. (52) means that the bending curvature tensor is constant restricted to Ω_1. This is a remarkable property of ellipses and can be used to solve inhomogeneous problems. The exterior solution to (51) is also explicitly given in Kellogg (1954).

7. Concluding comments

By judiciously using symmetry and heterogeneous materials, two simple mechanisms permit the engineering of a rather large electromechanical coupling in soft materials: electrets and flexoelectricity. Most importantly, the emergent macroscopic electromechanical coupling is reminiscent of piezoelectricity, i.e., the mechanical and electrical fields are

coupled even at the linear order; reversal of stimuli reverses the response and there is a true two-way coupling. While the electret mechanism is size-independent, flexoelectricity (as fully anticipated from past work) leads to a size-dependent response. What is surprising however is that, in soft materials, the impact of flexoelectricity is quite a bit stronger than even the highly flexoelectric crystalline ferroelectrics, and moreover, the effect is relevant even when the feature sizes are the order of microns. As shown in Section 4, a simple bilayer structure consisting of some rather typical polymers was shown to exhibit a piezoelectric response 20 times that of the well-known ferroelectric, Barium Titanate, at high enough stress. Although electrets have been experimentally exploited, albeit with little regard to their large-deformation behavior, soft flexoelectric composites of the type described in this work are yet to be made. Our linearized analytical expression (at least for electrets, (37)) clearly indicates that the reason for the large response in soft materials: the apparent piezoelectricity is inversely proportional to the difference in the stiffnesses of the constituents. Therefore, material heterogeneity is essential and soft materials will always result in a larger response. This can be carried even further by imaging a "foam composite": polymer consisting of voids. Although we did not analyze this configuration rigorously, use of our derived simple linear expression (which can be used for back-of-the-envelope calculation) indicates that we could potentially achieve upwards of 10 times Barium Titanate piezoelectricity (in the electret case) and many folds that for the flexoelectric case.

As has been well-studied by Suo and co-workers, e.g. Suo (2010), stability analysis is an important element of electromechanical response in soft materials. To our knowledge, no work has appeared so far in that direction pertaining to either electrets or flexoelectric materials. The latter, due to the presence of higher order terms requires some care and we expect this to be an interesting avenue for future research.

In regards to biological membranes, we have shown that in an otherwise electromechanically inert membrane, deformation can ensue due to a non-uniform applied field. For example, a simple point charge or ion can locally bend the membrane. We hope that some of the developments outlined in the present work will pave the way for studying the ramifications of charge-membrane interaction on hearing mechanism, ion transport and others—such an application is beyond the scope of the present work.

Acknowledgments

Appendix A. Derivation of Piola–Maxwell stress

The Piola–Maxwell stress (17) is the conjugate quantity of variation of deformation gradient for the electric energy. It turns out that the derivation of the expression of Maxwell stress in reference configuration is more transparent in a general three dimensional setting than the one dimensional setting in Section 2. This simply arises from the fact that direct tensor notations are much shorter than scalar notation.

To include the model in Section 2 as a special case, we consider a body $\Omega_R \subset \mathbb{R}^3$ in the *reference configuration*. Let Γ_D, Γ_N be a subdivision of the boundary $\partial\Omega_R$ with the electrostatic potential $\xi = \xi_b$ on Γ_R and surface charge density $\rho_{\text{surf}} = 0$ on Γ_N. Further, the body Ω contains an external polarization $\mathbf{P}_0^e : \Omega_R \to \mathbb{R}^3$, an intrinsic polarization $\mathbf{P} : \Omega_R \to \mathbb{R}^3$, and an external charge density $\rho_0^e : \Omega_R \to \mathbb{R}^3$. Let $\chi : \Omega_R \to \Omega$ be the deformation carrying every material point in the reference configuration to a spatial point in the current configuration Ω. As usual, a material point in the reference (current) configuration is denoted by Lagrangian coordinate $\mathbf{X} = (X, Y, Z)$ (Eulerian coordinate $\mathbf{x} = (x, y, z)$). The operators Grad, Div, Curl and grad, div, curl are taken with respect to Lagrangian coordinate $\mathbf{X}$ and Eulerian coordinate $\mathbf{x}$, respectively. Denote by $\mathbf{F} = \text{Grad}\,\chi$ the deformation gradient, $\mathbf{C} = \mathbf{F}^T\mathbf{F}$ the Cauchy–Green strain tensor and $J = \det \mathbf{F}$ the Jacobian. From the classic continuum mechanics, for any vector field $\mathbf{v} : \partial\Omega \to \mathbb{R}^3$ we have

$$\int_{\partial\Omega} \mathbf{v} \cdot \mathbf{n} \, ds = \int_{\partial\Omega_R} \mathbf{V} \cdot J\mathbf{F}^{-T}\mathbf{N} \, dS, \tag{A.1}$$

where $\mathbf{V} = \mathbf{v}(\chi(\mathbf{X}))$, and $\mathbf{n}$ (resp. $\mathbf{N}$) is the unit outward normal on the boundary of the body in the current (resp. reference) configuration.

By the Maxwell equation, we know that the electrostatic potential necessarily satisfies (cf., (4))

$$\begin{cases} \text{div}\,\mathbf{d} = \rho^e, \quad \mathbf{d} = -\varepsilon_0 \,\text{grad}\,\xi + \mathbf{p} + \mathbf{p}^e \quad \text{in } \Omega, \\ \xi = \xi_b \quad \text{on } \Gamma_D', \quad (-\varepsilon_0 \,\text{grad}\,\xi + \mathbf{p} + \mathbf{p}^e) \cdot \mathbf{n} = 0 \quad \text{on } \Gamma_N', \end{cases} \tag{A.2}$$

where $(\Gamma_D', \Gamma_N') = \chi(\Gamma_D, \Gamma_N)$ is the image of the boundaries, and (cf., (5))

$$(\mathbf{p}, \mathbf{p}^e, \rho^e) = \frac{1}{J}(\mathbf{P}, \mathbf{P}_0^e, \rho_0^e).$$

In the *reference configuration*, by (A.1) the above equation can be rewritten as

$$\begin{cases} \text{Div } \tilde{\mathbf{D}} = \rho_0^e, \qquad \tilde{\mathbf{D}} := -\varepsilon_0 J \mathbf{C}^{-1} \text{ Grad } \xi + \mathbf{F}^{-1}(\mathbf{P}+\mathbf{P}^e) & \text{in } \Omega_R, \\ \xi = \xi_b \quad \text{on } \Gamma_D, \quad \mathbf{N} \cdot \tilde{\mathbf{D}} = 0 \quad \text{on } \Gamma_R. \end{cases} \tag{A.3}$$

Upon solving the boundary value problem (A.2) or (A.3), we identity the electric energy as (cf., (8))

$$\mathcal{E}^{\text{elect}}[\chi, \mathbf{P}] = \frac{\varepsilon_0}{2} \int_\Omega |\text{grad } \xi|^2 + \int_{\Gamma_{D'}} \xi_b \mathbf{d} \cdot \mathbf{n} = \frac{\varepsilon_0}{2} \int_{\Omega_R} J |\mathbf{F}^{-T} \text{ Grad } \xi|^2 + \int_{\Gamma_D} \xi_b \tilde{\mathbf{D}} \cdot \mathbf{N}. \tag{A.4}$$

We now consider variations of deformation and polarization:

$$\chi \to \chi_\delta = \chi + \delta \chi_1, \quad \mathbf{P} \to \mathbf{P}_\delta = \mathbf{P} + \delta \mathbf{P}_1. \tag{A.5}$$

Algebraic calculations show that ($\mathbf{F}_1 = \text{Grad } \chi_1$)

$$\mathbf{F}_\delta^{-1} = \mathbf{F}^{-1} - \delta \mathbf{F}^{-1} \mathbf{F}_1 \mathbf{F}^{-1} + o(\delta), \quad J_\delta = J[1 + \delta \text{ Tr}(\mathbf{F}^{-1}\mathbf{F}_1)] + o(\delta), \mathbf{C}_\delta^{-1} = \mathbf{C}^{-1} - \delta(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T}) + o(\delta). \tag{A.6}$$

Further, by (A.3) we see that the electric potential ξ_δ satisfies

$$\begin{cases} \text{Div}[-\varepsilon_0 J_\delta \mathbf{C}_\delta^{-1} \text{ Grad } \xi_\delta + \mathbf{F}_\delta^{-1}(\mathbf{P} + \delta\mathbf{P}_1 + \mathbf{P}^e)] = \rho_0^e & \text{in } \Omega_R, \\ \xi_\delta - \xi_b = 0 \quad \text{on } \Gamma_D, \quad \tilde{\mathbf{D}}_\delta \cdot \mathbf{N} = 0 & \text{on } \Gamma_N. \end{cases} \tag{A.7}$$

From the above equation, it is easy to see that the actual electric field and hence the electric energy depend on the deformation. To find the change of electric field to the leading order, we assume that

$$\xi_\delta = \xi + \delta\xi_1 + o(\delta), \quad \tilde{\mathbf{D}}_\delta = \tilde{\mathbf{D}} + \delta\tilde{\mathbf{D}}_1 + o(\delta).$$

From the definition (A.3)$_1$ and (A.6), we find that

$$\tilde{\mathbf{D}}_1 = -\varepsilon_0 J \mathbf{C}^{-1} \text{ Grad } \xi_1 - \varepsilon_0 J \text{ Tr}(\mathbf{F}^{-1}\mathbf{F}_1)\mathbf{C}^{-1} \text{ Grad } \xi + \varepsilon_0 J(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T}) \text{ Grad } \xi - \mathbf{F}^{-1}\mathbf{F}_1\mathbf{F}^{-1}\mathbf{P} + \mathbf{F}^{-1}\mathbf{P}_1.$$

Inserting the above equation into (A.7) and keeping the terms of order δ, we obtain

$$\begin{cases} \text{Div } \tilde{\mathbf{D}}_1 = 0 & \text{in } \Omega_R, \\ \xi_1 = 0 \quad \text{on } \Gamma_D, \quad \tilde{\mathbf{D}}_1 \cdot \mathbf{N} = 0 \quad \text{on } \Gamma_N. \end{cases}$$

Multiplying the first of the above equations by ξ, by the divergence theorem we find that

$$\int_{\partial\Omega_R} \xi\tilde{\mathbf{D}}_1 \cdot \mathbf{N} - \int_{\Omega_R} \text{Grad } \xi \cdot \tilde{\mathbf{D}}_1 = 0. \tag{A.8}$$

That is,

$$\int_{\Omega_R} \varepsilon_0 J \text{ Grad } \xi \cdot \mathbf{C}^{-1} \text{ Grad } \xi_1 = -\int_{\partial\Omega_R} \xi\tilde{\mathbf{D}}_1 \cdot \mathbf{N} + \int_{\Omega_R} \text{Grad } \xi$$
$$\cdot [-\varepsilon_0 J \text{ Tr}(\mathbf{F}^{-1}\mathbf{F}_1)\mathbf{C}^{-1} \text{ Grad } \xi + \varepsilon_0 J(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T}) \text{ Grad } \xi - \mathbf{F}^{-1}\mathbf{F}_1\mathbf{F}^{-1}\mathbf{P} + \mathbf{F}^{-1}\mathbf{P}_1]. \tag{A.9}$$

Therefore,

$$\mathcal{E}^{\text{elect}}[\chi_\delta, \mathbf{P}_\delta] = \mathcal{E}^{\text{elect}}[\chi, \mathbf{P}] + \delta\text{Var1} + o(\delta), \tag{A.10}$$

where, by (A.4) and (A.6), we have

$$\text{Var1} = \frac{\varepsilon_0}{2} \int_{\Omega_R} \left[J \text{ Tr}\left(\mathbf{F}^{-1}\mathbf{F}_1\right)|\mathbf{F}^{-T} \text{ Grad } \xi|^2 - J \text{ Grad } \xi \cdot \left(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T}\right) \text{ Grad } \xi + 2J \text{ Grad } \xi \cdot \mathbf{C}^{-1} \text{ Grad } \xi_1 \right]$$
$$+ \int_{\Gamma_N} k\xi\xi_1 + \int_{\Gamma_D} \xi_b\tilde{\mathbf{D}}_1 \cdot \mathbf{N}.$$

Inserting (A.9) into the above equation we find that ($\mathbf{E} = -\mathbf{F}^{-T} \text{ Grad } \xi$)

$$\text{Var1} = \int_{\Omega_R} \left[-\frac{\varepsilon_0}{2} J \text{ Tr}\left(\mathbf{F}^{-1}\mathbf{F}_1\right)|\mathbf{E}|^2 + \frac{\varepsilon_0}{2} J\mathbf{E} \cdot \left(\mathbf{F}_1\mathbf{F}^{-1} + \mathbf{F}^{-T}\mathbf{F}_1^T\right)\mathbf{E} + \mathbf{E} \cdot \mathbf{F}_1\mathbf{F}^{-1}\mathbf{P} \right]$$
$$= \int_{\Omega_R} \left\{ \mathbf{F}_1 \cdot \left[-\frac{\varepsilon_0}{2} J|\mathbf{E}|^2\mathbf{F}^{-T} + \mathbf{E} \otimes \tilde{\mathbf{D}} - \mathbf{E} \cdot \mathbf{P}_1 \right] - \mathbf{E} \cdot \mathbf{P}_1 \right\} = \int_{\Omega_R} \left(\mathbf{F}_1 \cdot \tilde{\boldsymbol{\Sigma}}_{\text{MW}} - \mathbf{E} \cdot \mathbf{P}_1 \right), \tag{A.11}$$

where

$$\tilde{\boldsymbol{\Sigma}}_{\text{MW}} = -\frac{\varepsilon_0}{2} J|\mathbf{E}|^2\mathbf{F}^{-T} + \mathbf{E} \otimes \tilde{\mathbf{D}}$$

is the Piola–Maxwell stress. By (A.1), we recognize

$$\boldsymbol{\sigma}_{\mathrm{MW}} = \frac{1}{J}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}\mathbf{F}^T = (-\operatorname{grad}\xi) \otimes (-\varepsilon_0\operatorname{grad}\xi + \mathbf{p} + \mathbf{p}^e) - \frac{\varepsilon_0}{2}|\operatorname{grad}\xi|^2\mathbf{I} \tag{A.12}$$

is the familiar expression of Maxwell stress in the current configuration (Jackson, 1999).

We now come back to our setting in Section 2. From (3) we find that the deformation gradient is given by

$$\mathbf{F} = \begin{bmatrix} \lambda_1 & 0 & 0 \\ Y\alpha' & \lambda_2 & 0 \\ Z\beta' & 0 & \lambda_3 \end{bmatrix}.$$

Therefore, $\mathbf{F}^{-T}$ must be an upper triangle matrix of form ($*$ denotes possible non-zero components)

$$\mathbf{F}^{-T} = \begin{bmatrix} \lambda_1^{-1} & * & * \\ 0 & \lambda_2^{-1} & * \\ 0 & 0 & \lambda_3^{-1} \end{bmatrix},$$

and for variations in (10),

$$\mathbf{F}_1 = \begin{bmatrix} u_{1,X} & 0 & 0 \\ Y\alpha_{1,X} & \alpha_1 & 0 \\ Z\beta_{1,X} & 0 & \beta_1 \end{bmatrix}. \tag{A.13}$$

In Section 2 we have also assumed that the electric field, polarization and electric displacement are all in X-direction, and hence the Maxwell stress (A.12) in the current configuration is a diagonal matrix. Therefore, the *Piola–Maxwell* stress $\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} = J\boldsymbol{\sigma}_{\mathrm{MW}}\mathbf{F}^{-T}$ is necessarily an upper triangle matrix, and hence the inner product $\mathbf{F}_1 \cdot \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}$ has only contributions from diagonal components and is precisely given by (15)–(16).

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