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Emergent electromechanical coupling of electrets and some exact relations — The effective properties of soft materials with embedded external charges and dipoles

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ABSTRACT

Soft robotics, energy harvesting, large-deformation sensing and actuation, are just some of the applications that can be enabled by soft dielectrics that demonstrate substantive electromechanical coupling. Most soft dielectrics including elastomers, however, are not piezoelectric and rely on the universally present electrostriction and the Maxwell stress effect to enable the aforementioned applications. Electrostriction is a one-way electromechanical coupling and the induced elastic strain scales as ($\propto E^2$) upon the application of an electric field, E. The quadratic dependence of electrostriction on the electric field and the one-way coupling imply that, (i) A rather high voltage is required to induce appreciable strain, (ii) reversal of an applied bias will not reverse the sign of the deformation, and (iii) since it is a one-way coupling i.e. electrical stimuli may cause mechanical deformation but electricity *cannot* be generated by mechanical deformation, prospects for energy harvesting are rather difficult. An interesting approach for realizing an *apparent* piezoelectric-like behavior is to dope soft dielectrics with *immobile* charges and dipoles. Such materials, called electrets, are rather unique composites where a secondary material (in principle) is not necessary. Both experiments and supporting theoretical work have shown that soft electrets can exhibit a very large electromechanical coupling including a piezoelectric-like response. In this work, we present a homogenization theory for electret materials and provide, in addition to several general results, variational bounds and closed-form expressions for specific microstructures such as laminates and ellipsoidal inclusions. While we consider the nonlinear coupled problem, to make analytical progress, we work within the small-deformation setting. The specific conditions necessary to obtain a piezoelectric-like response and enhanced electrostriction are highlighted. There are very few universal, microstructure-independent *exact* results in the theory of composites. We succeed in establishing several such relations in the context of electrets.

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1. Introduction

The simple act of a robotic arm lifting an object requires the underlying material to be capable of large mechanical deformations and that it deform in response to a stimulus with moderate energetic cost. This example epitomizes the imperative to develop soft materials that exhibit strong electromechanical coupling. In addition to robotics, electromechanical soft materials have exciting applications in many other contexts also: energy harvesting, stretchable electronics, prostheses, nano-electromechanical-systems and others cf. Carpi et al. (2008), Carpi et al. (2010), Erturk and Inman (2011), Xu (2010), Bauer et al. (2014), Rogers et al. (2010), Huang et al. (2013), Yang et al., Muralt et al. (2009), Trolier-McKinstry and Muralt (2004), Long et al. (2005).

The *ideal* electromechanical coupling is the well-known phenomenon of piezoelectricity which is the linear coupling between electrical fields and mechanical deformation. Piezoelectrics can be actuated by a small voltage, mechanical motion can be converted into electricity making energy harvesting possible and the reversal of stimuli (whether mechanical or electrical) leads to a reversal of the response (respectively electrical or mechanical). Unfortunately, only a rather limited set of materials—mostly *hard* and crystalline—possess this remarkable property. Examples include materials like barium titanate and lead zirconium titanate. Such piezoelectrics, with elastic moduli in the order of 100's of GPa, are all but useless for applications that require large deformations. Polymers like polyvinylidene difuoride (PVDF) are among the few polymers that exhibit piezoelectricity (Murayama et al., 1976) but the effect is rather weak and PVDF is not quite *as* soft as some of the applications would necessitate.

Dielectric elastomers are, at the moment, arguably the most intensely researched class of materials as far as soft electromechanical materials are concerned. Upon application of a suitably high voltage, an area increase of nearly 1700% has recently been demonstrated for an acrylic membrane (Keplinger et al., 2012). Indeed, through clever design, dielectric elastomers have been successfully used for myriad applications like braille displays, adaptive optics, and energy harvesting (cf. Bauer et al., 2014; Huang et al., 2013; Keplinger et al., 2012; Koh et al., 2009; Shankar et al., 2007; Trivedi et al., 2008; Yang et al., 0000). What is notable however is that dielectric elastomers are not piezoelectric. Their electromechanical coupling emerges entirely from an effect that is universally present in all dielectrics — the so-called Maxwell stress effect and/or electrostriction[1]. In these phenomena, an electric field exerts a force on all dielectrics proportional to E^2 where E is the applied electric field. This force is small and accordingly, the induced deformation in hard dielectrics is negligible[2]. This scenario changes drastically for soft dielectric elastomers; as demonstrated convincingly in several works (Keplinger et al., 2012; Li et al., 2004; Pelrine et al., 2000). There are some drawbacks however. Electrostriction is a one-way coupling. An electric field will produce deformation but a mechanical stress, in its naive form of application, will not coax any electricity from the material—unlike piezoelectric materials. The absence of this direct converse effect implies that energy harvesting is rather difficult[3]. Furthermore, the quadratic dependence of the deformation on electric fields implies that upon reversal of the applied voltage, the deformation will not reverse and very high voltages are required for actuation.

The development of another class of materials — the so-called electrets — appears to be a promising route to create soft materials that are piezoelectric-like, capable of large deformation[4] and may be prospects for engineering high electromechanical coupling. Electrets are, simply put, dielectrics that have trapped "quasi-permanent" immobile charges and/or dipoles[5]. Although the "modern" discovery of electrets dates back to the work by the Japanese scientist, Mototaro Eguchi in 1919, as far as the authors are aware, this field received new life in the eighties by groups in Germany at the hands of research groups (Gerhard-Multhaupt, Bauer et al., 2004; Buchberger et al., 2008; Sessler, 1987; Sessler and Hillenbrand, 1999; Wegener and Bauer, 2005) led by Sessler, Gerhard-Multhaupt, Gross and Bauer[6]. Perhaps the most striking example of an electret material is the creation of polypropylene foams—which have a distinct cellular structure characterized by the presence of micron sized voids (Fig. 1). Charges are embedded (and trapped) on the void surface via a process that leads to electrical breakdown of the air in the voids. Apparent piezoelectric coefficient as high as 1200 pC/N have been reported (Hillenbrand and Sessler, 2008) for such optimally fabricated soft foams—which is six times that of the well-known piezoelectric, lead zirconium titanate.

What is the principle underpinning the apparent piezoelectricity of electrets? The basic notion can be explained in a rather simple manner by considering a bilayer film structure as shown in Fig. 2. Consider a quasi-permanent immobile

[1] Strictly speaking, electrostriction and the Maxwell-stress effect are physically distinct, however, mathematically similar (Tian, 2007; Tian et al., 2012; Zhao and Suo, 2008). In what follows, we will simply use the terminology "electrostriction" and it is to be understood that both effects can be lumped together.

[2] For example, under similar conditions as used for the acrylic dielectric elastomer membrane that led to a 1700% areal change, the well-known hard dielectric, silicon, will barely change its area by 0.001%

[3] We remark that there are some exceptional works by Bauer, Koh, Zhao, Suo, among other (cf. Koh et al., 2011, 2009) who, using certain design are able to scavenge electricity from elastomers. In one instance, the demonstration involves harvesting from ocean waves.

[4] Unfortunately, not to the level of elastomers. Stabilizing charges and dipoles in elastomers for a usable time-period remains to be an open experimental challenge

[5] By immobile we mean that embedded charges and dipoles only shift position due to deformation. In the time-period of interest or observation, the charges do not flow or move and are thus "quasi-permanent"; a term coined by the early pioneers in electret research. The word "quasi" is used to indicate that an electret material is in a metastable state. The charges have a driving force to "flow" and leak away but due to the presence of interfaces or trapping states, are locked in place for an appreciable amount of time.

[6] A nice historical account leading up to the eighties may be found in a paper by Gerhard-Multhaupt and the book by Sessler (1987).

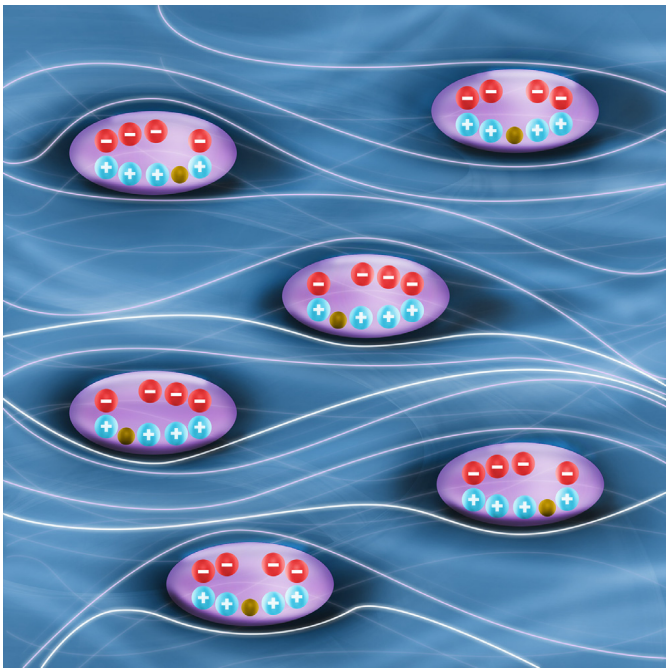

Fig. 1. A schematic of an electret based on polymer foam with embedded dipoles on the void surfaces. This artistic rendition is inspired from a figure in Bauer et al. (2004).

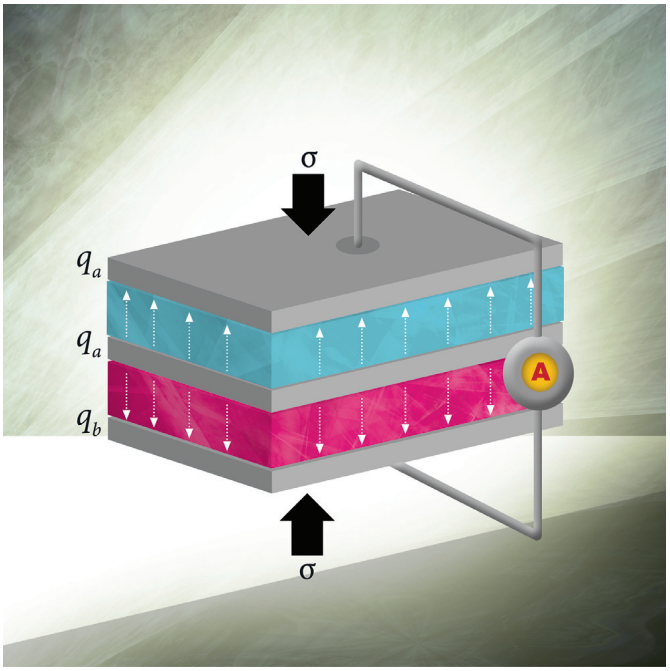

Fig. 2. The basic principle underlying the apparent piezoelectric behavior of electrets.

charge layer q embedded at the interface between the two different soft elastic materials. There will exist, due to the presences of this charge, a state of residual electrical field as well as induced charges q_a and q_b at the opposite electrodes. Due to electrostriction, even prior to an external mechanical or electrical load, a thickness change $\Delta h \sim q^2$ is expected. Therefore, in addition to a residual electrical field, there also exists a state of residual strain and stress. If now, an external stress is applied to this electret construction, the resulting deformation will convect the existing electric field state in the body and change the induced charges at the electrodes. This resulting change in electric field can be shown to be linearly proportional to the applied stress and appears therefore as an emergent piezoelectric effect. A similar situation ensues upon the application of an applied voltage difference, V. In that case, the total thickness change scales as: $\Delta h \sim a \cdot q^2 + b \cdot V^2 + c \cdot$

qV where (a, b, c) are geometry and material-dependent constants. The first term, as indicated earlier, is due to the Maxwell stress caused by the electrical field due to the embedded charge (q), the second term is the deformation due to the Maxwell stress caused by the applied voltage, V and finally, a term, linear in V, emerges due to the interaction between the applied field and the presence of charge q. This linear term in V manifests as an apparent converse piezoelectric effect (—and in fact, cq can be shown to be proportional to the piezoelectic constant).

Surprisingly, relatively fewer modeling efforts have been dedicated to electrets. Simple (but certainly insightful) formulae on the lines of what has been described in the preceding paragraph pertaining to simple bilayer structures have been long known (Kacprzyk et al., 1995). More recently, Deng et al. (2014b), Deng et al. (2014a) have provided systematic (nonlinear) solutions to boundary value problems such as the one shown in Fig. 2. In particular, the linearized solution while different from the simpler estimates in the literature, confirm the basic scaling. However, that being said, solutions to bilayers such as in Fig. 2, while useful for illustrative purposes, do not present the *homogenization* solution for the calculation of bulk emergent properties of macroscopic electret materials[7]. The homogenization of electret materials appears to have received scant attention in the literature — with the notable (and perhaps sole) exception of the work by Lefevre and Lopez-Pamies (2017). In this work, using a variational approach and two-scale asymptotics, we present a homogenization theory for soft electret materials and establish general relations for the emergent piezoelectric and electrostrictive response. Specifically, we provide insights into the conditions under which apparent piezoelectricity may be expected from an electret material. To make decisive progress, using a scaling established by Tian, Bhattacharya and co-workers (Tian, 2007; Tian et al., 2012), we work in a linearized setting although remark that the coupled problem is nonlinear. We present variational bounds and closed-form expressions for specific microstructures such as laminates and ellipsoidal inclusions and finally establish some, in our view, remarkable and unexpected exact results.

The outline of the paper is as follows: in Section 2, we summarize a nonlinear continuum theory for electrostatics of deformable media and outline the linearization assumptions that will be adopted in the present work. While, the central goal of the our work is to derive the homogenized piezoelectric response of electrets, the corresponding consideration of electrostriction is a necessary route to such a calculation as piezoelectricity in electrets emerges from the interaction of electrostriction and pre-existing residual electrical and mechanical fields. Accordingly, Section 3 is devoted to the homogenization of electrostriction where we have re-defined the effective electrostrictive tensor in Tian (2007), Tian et al. (2012) by a variational principle. We address the effective electro-elastic properties and homogenization of electrets in Section 4 by the standard method of two-scale convergence (Allaire, 1992; Cioranescu and Donato, 1999), and remark that Ref. Lefevre and Lopez-Pamies (2017) considered a more general system that includes the effects of "active charges". We focus specifically on the piezoelectric response in Section 5. In Section 6, we present variational bounds, some exact relations between effective electro-elastic properties, and closed-form results for specific microstructures such as laminates and ellipsoidal inclusions.

2. A summary of a nonlinear continuum theory for electrostatics of deformable materials

In this section, we briefly summarize the continuum theory we will use to describe the coupled electromechanical behavior of electrets. The electro-elastic theory for a continuum body has been the theme of a number of earlier works including Toupin (1956), Eringen and Maugin (1990), Dorfmann and Ogden (2005), Suo and co-workers (Suo et al., 2008), Suo (2010), McMeeking and Landis (2005), and Xiao and Bhattacharya (2008). The polarization-based formulation and the systematic linearization of constitutive relations have been addressed by Tian and co-workers (Tian, 2007; Tian et al., 2012). A recent exposition which compares various flavors of electro-elastic theories can be found in the two papers by Liu (2013), Liu (2014). Below we briefly outline the general nonlinear theory, clarify the assumptions for a linearized theory of electrostriction, and derive the implications of material symmetry on the electrostrictive tensor.

Notation. For brevity, wherever possible, we employ direct notation. Vectors are denoted by bold symbols such as **e, u**, etc. When index notation is used, the convention of summation over repeated indices is followed. The tensor product between two vectors $\mathbf{a}, \mathbf{b} \in \mathbb{R}^3$ is defined as $(\mathbf{a} \otimes \mathbf{b})_{ij} = (\mathbf{a})_i(\mathbf{b})_j$ whereas the inner (or dot) product is defined as $\langle \mathbf{a}, \mathbf{b} \rangle \equiv \mathbf{a} \cdot \mathbf{b} := (\mathbf{a})_i(\mathbf{b})_i$, and the inner (or dot) product between matrices $\mathbf{A}$ and $\mathbf{B}$ of the same size is defined as $\mathbf{A} \cdot \mathbf{B} := \mathrm{Tr}(\mathbf{A}^T\mathbf{B}) = (\mathbf{A})_{ij}(\mathbf{B})_{ij}$. For a domain D, $f_D(\,)$ denote the average of the integrand on D. From the viewpoint of matrices, the ith row vector of the gradient of a vector field, e.g., $\nabla\mathbf{u}$, is the gradient of the ith component of $\mathbf{u}$ (with respect the Lagrangian coordinates $\mathbf{x}$, unless stated otherwise) whereas the "div" operates on the row vectors of a matrix field. Therefore, $\mathrm{div}\nabla\mathbf{u} = \Delta\mathbf{u}$ and $\mathrm{div}[(\nabla\mathbf{u})^T] = \nabla(\mathrm{div}\mathbf{u})$. For a scaling parameter $0 < \varepsilon \ll 1$ and a real number n, $O(\varepsilon^n)$ implies the asymptotic behavior $O(\varepsilon^n)/\varepsilon^n \to C \neq 0$ as $\varepsilon \to 0$ where as $o(\varepsilon^n)/\varepsilon^n \to 0$ as $\varepsilon \to 0$.

Consider a deformable dielectric body with reference configuration $D \subset \mathbb{R}^3$. To adequately account for electret materials we will need to consider the interaction of embedded charges and dipoles with electrostriction and therefore those aspects are emphasized in the outline of the continuum theory. We begin with the kinematics and assume that the thermodynamic state of the body is described by the deformation and nominal polarization $(\mathbf{y}, \mathbf{p}) : D \to \mathbb{R}^3 \times \mathbb{R}^3$. By the Maxwell equations, we introduce the electric potential ξ, and for simplicity, consider the Dirichlet boundary conditions:

$$\xi = \xi_b \quad \text{on } \partial D \qquad \text{and} \qquad \mathbf{y} = \mathbf{x} + \mathbf{u}_b \quad \text{on } \partial D, \tag{1}$$

[7] In due course, in this work, we will carefully distinguish between what we have termed as "apparent" piezoelectricity and "true effective piezoelectricity".

where $\xi_b : \partial D \to \mathbb{R}$ (resp. $\mathbf{u}_b : \partial D \to \mathbb{R}^3$) is the imposed boundary potential (resp. boundary displacement). As usual, we denote by

$$\mathbf{F} = \nabla \mathbf{y}, \quad J = \det \mathbf{F}, \quad \mathbf{C} = \mathbf{F}^T \mathbf{F}$$

the deformation gradient, Jacobian, and Cauchy–Green tensor, respectively.

The nominal polarization $\mathbf{p}$ represents the dipole density per unit volume in the reference configuration. Therefore, the Maxwell equation can be written as (ϵ_0 is the vacuum permittivity)

$$\mathrm{div}_{\mathbf{y}}(-\epsilon_0 \nabla_{\mathbf{y}} \xi + \mathbf{p}/J) = 0 \qquad \text{in } \mathbf{y}(D), \tag{2}$$

where subscript $_{\mathbf{y}}$ means that the derivatives are taken with respect to the Eulerian coordinates $\mathbf{y}$. In what follows, to conveniently couple deformation to electrical quantities, it is expedient to work in the *reference* configuration where the Maxwell's Eq. (2) is transformed to:

$$\nabla \cdot (-\epsilon_0 J \mathbf{C}^{-1} \nabla \xi + \mathbf{F}^{-1} \mathbf{p}) = 0 \qquad \text{in } D, \tag{3}$$

where (for now) we have assumed that there are no external charges or dipoles.

The free energy of the system is postulated as Tian (2007), Liu (2014):

$$F[\mathbf{y}, \mathbf{p}] = \int_D \left[\Psi(\nabla \mathbf{y}, \mathbf{p}) + \frac{\epsilon_0}{2} \nabla \xi \cdot J \mathbf{C}^{-1} \nabla \xi \right] d\mathbf{x} + \int_{\partial D} \xi_b \mathbf{n} \cdot (-\epsilon_0 J \mathbf{C}^{-1} \nabla \xi + \mathbf{F}^{-1} \mathbf{p}) d\mathbf{x}, \tag{4}$$

where the internal energy density function $\Psi : \mathbb{R}^{3\times 3} \times \mathbb{R}^3 \to \mathbb{R}$ prescribes the electro-elastic constitutive laws of the material, the second term is equivalent to the electric field energy $\int_{\mathbf{y}(D)} \frac{\epsilon_0}{2} |\nabla_{\mathbf{y}} \xi|^2$, and the last term is the potential energy associated with the electric device (e.g., the battery) used for maintaining the boundary potential $(1)_1$. By (3) and the divergence theorem, we may rewrite the free energy functional (4) as

$$F[\mathbf{y}, \mathbf{p}] = \int_D \left[\Psi(\nabla \mathbf{y}, \mathbf{p}) - \frac{\epsilon_0}{2} \nabla \xi \cdot J \mathbf{C}^{-1} \nabla \xi + \nabla \xi \cdot \mathbf{F}^{-1} \mathbf{p} \right] d\mathbf{x}. \tag{5}$$

From the principle of minimum free energy, the equilibrium state of the body is determined by the variational problem (ξ is determined by (3) and $(1)_1$):

$$\min_{(\mathbf{y}, \mathbf{p})} \left\{ F[\mathbf{y}, \mathbf{p}] : \mathbf{y} \text{ satisfies } (1)_2 \text{ and } \int_D |\mathbf{p}|^2 < +\infty \right\}. \tag{6}$$

The Euler–Lagrange equations associated with (6) are given by Liu (2014)

$$\begin{cases} \mathbf{F}^{-T} \nabla \xi + \frac{\partial \Psi}{\partial \mathbf{p}} = 0 & \text{in } D, \\ \mathrm{div}\left(\frac{\partial \Psi(\nabla \mathbf{y}, \mathbf{p})}{\partial \nabla \mathbf{y}} + \mathbf{\Sigma}_{\mathrm{MW}} \right) = 0 & \text{in } D, \end{cases} \tag{7}$$

where $\mathbf{\Sigma}_{\mathrm{MW}}$ denotes the Piola–Maxwell stress and is given by

$$\mathbf{\Sigma}_{\mathrm{MW}} = -\frac{\epsilon_0}{2} J (\nabla \xi \cdot \mathbf{C}^{-1} \nabla \xi) \mathbf{F}^{-T} + (\mathbf{F}^{-T} \nabla \xi) \otimes (-\epsilon_0 J \mathbf{C}^{-1} \nabla \xi + \mathbf{F}^{-1} \mathbf{p}). \tag{8}$$

We remark that (7), together with the Maxwell Eq. (3) and boundary conditions (1), form a closed system of nonlinear differential equations for ($\mathbf{y}$, $\mathbf{p}$, ξ), and would presumably admit solutions reflecting the electro-mechanical behavior of the material.

For a simplified theory of electrostriction, we choose the natural state of the body as the reference configuration, meaning that ($\mathbf{I}$ is the identity matrix in $\mathbb{R}^{3\times 3}$)

$$\left. \frac{\partial \Psi}{\partial \mathbf{F}} \right|_{(\mathbf{F}, \mathbf{p}) = (\mathbf{I}, 0)} = \left. \frac{\partial \Psi}{\partial \mathbf{p}} \right|_{(\mathbf{F}, \mathbf{p}) = (\mathbf{I}, 0)} = 0. \tag{9}$$

Also, we assume the material is *intrinsically* non-piezoelectric in the sense that

$$\left. \frac{\partial^2 \Psi}{\partial \mathbf{F} \partial \mathbf{p}} \right|_{(\mathbf{F}, \mathbf{p}) = (\mathbf{I}, 0)} = 0. \tag{10}$$

Third, we restrict ourselves to the regime of small deformation and moderately small electric field ($\mathbf{u}(\mathbf{x}) = \mathbf{y}(\mathbf{x}) - \mathbf{x}$ is the displacement):

$$\nabla \mathbf{u} \sim \varepsilon \ll 1, \qquad \mathbf{p} \sim \varepsilon^{1/2} \text{ (and hence } -\nabla \xi \sim \varepsilon^{1/2} \text{ by (3))}. \tag{11}$$

Then, as shown by Tian and co-workers Tian (2007), Tian et al. (2012) and Liu (2014) we can formally decompose the free energy (5) according to their order of magnitude as compared with the small parameter ε. To this end, by (11) we recall the algebraic identities

$$\begin{aligned} \mathbf{F}^{-1} &= (\mathbf{I} + \nabla \mathbf{u})^{-1} = \mathbf{I} - \nabla \mathbf{u} + O(\varepsilon^2), \\ J &= 1 + \mathbf{I} \cdot \nabla \mathbf{u} + O(\varepsilon^2), \end{aligned}$$

$$\mathbf{C}^{-1} = \mathbf{I} - \nabla\mathbf{u} - (\nabla\mathbf{u})^T + O(\varepsilon^2),$$
$$J\mathbf{C}^{-1} = \mathbf{I} + (\nabla\cdot\mathbf{u})\mathbf{I} - \nabla\mathbf{u} - (\nabla\mathbf{u})^T + O(\varepsilon^2).$$

Inserting the above equations into (4), by (11) we obtain

$$F[\mathbf{y},\mathbf{p}] = F^{(0)} + F^{(1)} + F^{(2)} + o(\varepsilon^2), \tag{12}$$

where the first term $F^{(0)} := F[\mathbf{y}=\mathbf{x},\mathbf{p}=0]$ is independent of the state variables (**u, p**),

$$F^{(1)}[\mathbf{p}] = \int_D \left[\frac{1}{2}\mathbf{p}\cdot\boldsymbol{\chi}\mathbf{p} + \frac{\epsilon_0}{2}|\nabla\xi|^2\right]d\mathbf{x} + \int_{\partial D}\xi_b\mathbf{n}\cdot(-\epsilon_0\nabla\xi+\mathbf{p})d\mathbf{x} \sim \varepsilon,$$
$$F^{(2)}[\mathbf{u},\mathbf{p}] = \int_D \left[\frac{1}{2}\nabla\mathbf{u}\cdot\mathbb{C}\nabla\mathbf{u} + \nabla\mathbf{u}\cdot\mathbb{M}(\mathbf{p}\otimes\mathbf{p}) + \nabla\mathbf{u}\cdot\boldsymbol{\sigma}_{\mathrm{MW}}\right]d\mathbf{x} \sim \varepsilon^2. \tag{13}$$

Also, by (8) the (leading-order) Maxwell stress is given by

$$\boldsymbol{\Sigma}_{\mathrm{MW}} = \boldsymbol{\sigma}_{\mathrm{MW}} + o(\varepsilon), \qquad \boldsymbol{\sigma}_{\mathrm{MW}} = -\frac{\epsilon_0}{2}|\nabla\xi|^2\mathbf{I} + \epsilon_0\nabla\xi\otimes\nabla\xi - \nabla\xi\otimes\mathbf{p}, \tag{14}$$

and the tensors $\boldsymbol{\chi}, \mathbb{C}, \mathbb{M}$ are defined as (all derivatives are evaluated at $(\mathbf{F},\mathbf{p}) = (\mathbf{I},0)$)

$$\boldsymbol{\chi} = \frac{\partial^2\Psi}{\partial\mathbf{p}\partial\mathbf{p}}, \quad \mathbb{C} = \frac{\partial^2\Psi}{\partial\mathbf{F}\partial\mathbf{F}}, \quad \mathbb{M} = \frac{1}{2}\frac{\partial^3\Psi}{\partial\mathbf{F}\partial\mathbf{p}\partial\mathbf{p}}. \tag{15}$$

Similarly, we can rewrite Eq. (3) as

$$\nabla\cdot(-\epsilon_0\nabla\xi+\mathbf{p}) + o(\varepsilon^{1/2}) = 0 \quad \text{in } D. \tag{16}$$

Then the variational principle (6), together with the constraint (3), implies the sequential minimization problems

$$\min_{\mathbf{p}}\{F^{(1)}[\mathbf{p}] : \xi \text{ satisfies } (1)_1 \text{ and } (16)\}, \text{ and}$$
$$\min_{\mathbf{u}}\{F^{(2)}[\mathbf{u},\mathbf{p}] : \mathbf{u} \text{ satisfies } (1)_2\}. \tag{17}$$

Immediately, we find that the associated Euler–Lagrange equations for (**p, u**) are given by

$$\begin{cases}\nabla\xi + \boldsymbol{\chi}\mathbf{p} = 0,\\ \mathrm{div}[\mathbb{C}\nabla\mathbf{u} + \mathbb{M}(\mathbf{p}\otimes\mathbf{p}) + \boldsymbol{\sigma}_{\mathrm{MW}}] = 0.\end{cases} \tag{18}$$

Taking into account (16) to the leading order and eliminating **p** by $(18)_1$, we conclude the following system of differential equations for (**u**, ξ):

$$\begin{cases}\nabla\cdot(\boldsymbol{\epsilon}\nabla\xi) = 0 & \text{in } D,\\ \mathrm{div}[\mathbb{C}\nabla\mathbf{u} + \mathbb{A}(\nabla\xi\otimes\nabla\xi)] = 0 & \text{in } D,\end{cases} \tag{19}$$

where $\boldsymbol{\epsilon} = \epsilon_0\mathbf{I} + \boldsymbol{\chi}^{-1}$,

$$(\mathbb{A})_{ijkl} = (\mathbb{M})_{ijk'l'}(\boldsymbol{\chi}^{-1})_{kk'}(\boldsymbol{\chi}^{-1})_{ll'} + \frac{\epsilon_0}{2}\mathbb{T}_{ijkl} + \frac{1}{2}[\delta_{ik}(\boldsymbol{\chi}^{-1})_{jl} + \delta_{il}(\boldsymbol{\chi}^{-1})_{jk}], \tag{20}$$

and the fourth-order tensor $\mathbb{T}:\mathbb{R}^{3\times3}\to\mathbb{R}^{3\times3}$ is defined as

$$\mathbb{T}\mathbf{F} = \mathbf{F} + \mathbf{F}^T - (\mathrm{Tr}\mathbf{F})\mathbf{I} \qquad \forall\,\mathbf{F}\in\mathbb{R}^{3\times3},$$
$$\mathbb{T}_{ijkl} = \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \delta_{ij}\delta_{kl}. \tag{21}$$

The following aspects are notable:

- We recognize the tensor $\boldsymbol{\epsilon}\in\mathbb{R}^{3\times3}_{\mathrm{sym}}$ as the *permittivity or dielectric* tensor and refer to the fourth-order tensor $\mathbb{A}$ (or $\mathbb{M}$) as the *electrostrictive* tensor.
- The scaling introduced in (11) enables us to go from a fully coupled nonlinear system of equations to a semi-coupled linear one. That is, we may now first solve $(19)_1$ for the electrical quantities without regard for deformation, and those then enter as "forcing" terms for the mechanical equilibrium equations $(19)_2$.
- The system (19), together with the boundary conditions (1), forms a boundary value problem for (**u**, ξ) whose existence, uniqueness and regularity can be addressed by the classical theory for elliptic systems (Evans, 2010).
- The sequential variational problem (17) obtained by the above formal calculations can be rigorously justified as the asymptotic limit of (6) by the Γ-convergence method, see Tian (2007), Tian et al. (2012). To summarize, in the regime of small deformation and moderately small electric field and at the absence of intrinsic piezoelectricity (i.e. (10)), the leading electro-mechanical coupling arises from the electrostriction and is governed by the variational principle (17) or the associated Euler–Lagrange equations (19).

Remark 1. The principles of frame indifference and material symmetries imply that the internal energy density function $\Psi = \Psi(\mathbf{F}, \mathbf{p})$ has to satisfy

$$\begin{cases} \Psi(\mathbf{RF}, \mathbf{Rp}) = \Psi(\mathbf{F}, \mathbf{p}) & \forall\, \mathbf{R} \in SO(3), \\ \Psi(\mathbf{FQ}, \mathbf{p}) = \Psi(\mathbf{F}, \mathbf{p}) & \forall\, \mathbf{Q} \in \mathcal{G}, \end{cases} \tag{22}$$

where $\mathcal{G} \subset SO(3)$ is the point group associated with the material symmetries. It is well-known that (22) places nontrivial restrictions on the material property tensors appeared in a linearized theory. In present context, we are particularly interested in the restrictions on the electristrictive tensor $\mathbb{A}$ (cf., (20)) or $\mathbb{M}$ (cf., (15)) since the restrictions on $\mathbb{C}$ (resp. $\boldsymbol{\chi}$) can be obtained by setting $\mathbf{p} = 0$ (resp. $\mathbf{F} = \mathbf{I}$) in (22) and have been addressed in standard textbooks (e.g., Gurtin et al., 2010).

First, we consider an infinitesimal rigid rotation with $\mathbf{R} = \mathbf{I} + \mathbf{W}$ and $\mathbb{R}^{3\times3}_{\text{skew}} := \{\mathbf{M} \in \mathbb{R}^{3\times3} : \mathbf{M}^T = -\mathbf{M}\} \ni \mathbf{W} \sim \varepsilon$. By $(22)_1$ and (11) we find that

$$(H_{ij} + W_{ij})\mathbb{M}_{ijkl} p_k p_l + W_{ij} p_j \chi_{il} p_l = H_{ij}\mathbb{M}_{ijkl} p_k p_l$$

for all $\mathbf{H} \in \mathbb{R}^{3\times3}$, $\mathbf{W} \in \mathbb{R}^{3\times3}_{\text{skew}}$, $\mathbf{p} \in \mathbb{R}^3$, and hence

$$\mathbb{M}_{ijkl} + \chi_{il}\delta_{jk} = \mathbb{M}_{jikl} + \chi_{jl}\delta_{ik}, \qquad \mathbb{M}_{ijkl} = \mathbb{M}_{ijlk}, \tag{23}$$

where the last equality follows from the definition of $\mathbb{M}$-tensor $(15)_3$. Moreover, by (22) we have

$$\Psi(\mathbf{F}, \mathbf{p}) = \Psi(\mathbf{RF}, \mathbf{Rp}) = \Psi(\mathbf{RFQ}, \mathbf{Rp}) = \Psi(\mathbf{Q}^T\mathbf{FQ}, \mathbf{Q}^T\mathbf{p}) \qquad \forall\, \mathbf{Q} \in \mathcal{G},$$

where the second equality follows from $(22)_2$ and the last follows by setting $\mathbf{R} = \mathbf{Q}^T$. In account of the classic restrictions on $\mathbb{C}$ and $\boldsymbol{\chi}$, by (11) we conclude that

$$\mathbb{M}_{i'j'k'l'} Q_{ii'} Q_{jj'} Q_{kk'} Q_{ll'} = \mathbb{M}_{ijkl} \quad \forall\, \mathbf{Q} \in \mathcal{G}. \tag{24}$$

From (23) and (24) we observe that as compared with the elasticity tensor $\mathbb{C}$, the electrostrictive tensor $\mathbb{M}$ does not enjoy the minor or major symmetry i.e. in general, $\mathbb{M}_{ijkl} \neq \mathbb{M}_{jikl}$ and $\mathbb{M}_{ijkl} \neq \mathbb{M}_{klij}$. However, for isotropic materials with $\mathcal{G} = SO(3)$, the electrostrictive tensor $\mathbb{M}$ has to be of the same form as an isotropic elasticity tensor and can be written as

$$\mathbb{M}_{ijkl} = m_\mu(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) + m_\lambda \delta_{ij}\delta_{kl},$$

where m_μ, m_λ are two material constants in analogy with the Lamé constants of an isotropic elastic material.

Remark 2. For an ideal dielectric material whose electric permittivity tensor $\boldsymbol{\epsilon}$ is independent of deformation, the internal energy density function $\Psi = \Psi(\mathbf{F}, \mathbf{p})$ has to be of the following form (Liu, 2014):

$$\Psi(\mathbf{F}, \mathbf{p}) = W(\mathbf{U}) + \frac{1}{2J}\mathbf{p} \cdot \mathbf{R}\boldsymbol{\chi}\mathbf{R}^T\mathbf{p},$$

where $W = W(\mathbf{U})$ is the elastic energy density, $\mathbf{U} = \mathbf{C}^{1/2}$, $\boldsymbol{\chi} = (\boldsymbol{\epsilon} - \epsilon_0\mathbf{I})^{-1}$, and $\mathbf{R} = \mathbf{FU}^{-1} \in SO(3)$. To calculate the tensor $\mathbb{M}$ defined by (15), we expand the scalar function $(\varepsilon, \eta) \mapsto \Psi(\mathbf{I} + \varepsilon\mathbf{F}_1, \eta\mathbf{p}_1)$ for fixed $(\mathbf{F}_1, \mathbf{p}_1) \in \mathbb{R}^{3\times3} \times \mathbb{R}^3$ at $(\varepsilon, \eta) = 0$:

$$\Psi(\mathbf{I} + \varepsilon\mathbf{F}_1, \eta\mathbf{p}_1) = W(\mathbf{U}_\varepsilon) + \eta^2\frac{1}{2}\mathbf{p}_1 \cdot \boldsymbol{\chi}\mathbf{p}_1 + \frac{\eta^2\varepsilon}{2}[-\mathbf{p}_1 \cdot \boldsymbol{\chi}\mathbf{p}_1 \mathrm{Tr}\mathbf{F}_1 - \mathbf{F}_1 \cdot (\boldsymbol{\chi}\mathbf{p}_1) \otimes \mathbf{p}_1 + \mathbf{F}_1^T \cdot (\boldsymbol{\chi}\mathbf{p}_1) \otimes \mathbf{p}_1] + o(\eta^2\varepsilon),$$

where we have employed the following algebraic identities:

$$\begin{aligned} \mathbf{C}_\varepsilon &= (\mathbf{I} + \varepsilon\mathbf{F}_1)^T(\mathbf{I} + \varepsilon\mathbf{F}_1) = \mathbf{I} + \varepsilon(\mathbf{F}_1 + \mathbf{F}_1^T) + o(\varepsilon), \\ \mathbf{U}_\varepsilon^{-1} &= \mathbf{C}_\varepsilon^{-1/2} = \mathbf{I} - \frac{\varepsilon}{2}(\mathbf{F}_1 + \mathbf{F}_1^T) + o(\varepsilon), \\ \mathbf{R}_\varepsilon &= (\mathbf{I} + \varepsilon\mathbf{F}_1)\mathbf{U}_\varepsilon^{-1} = \mathbf{I} + \frac{\varepsilon}{2}(\mathbf{F}_1 - \mathbf{F}_1^T) + o(\varepsilon), \\ J_\varepsilon^{-1} &= 1/\det(\mathbf{I} + \varepsilon\mathbf{F}_1) = 1 - \varepsilon\mathrm{Tr}\mathbf{F}_1 + o(\varepsilon), \\ \mathbf{p}_1 \cdot \mathbf{R}_\varepsilon\boldsymbol{\chi}\mathbf{R}_\varepsilon^T\mathbf{p}_1 &= \frac{\varepsilon}{2}\mathbf{F}_1 \cdot [\mathbf{p}_1 \otimes (\boldsymbol{\chi}\mathbf{p}_1) - (\boldsymbol{\chi}\mathbf{p}_1) \otimes \mathbf{p}_1] + o(\varepsilon). \end{aligned}$$

Therefore,

$$\mathbf{F}_1 \cdot \mathbb{M}(\mathbf{p}_1 \otimes \mathbf{p}_1) = \frac{1}{2}\mathbf{F}_1 \cdot [\mathbf{p}_1 \otimes (\boldsymbol{\chi}\mathbf{p}_1) - (\boldsymbol{\chi}\mathbf{p}_1) \otimes \mathbf{p}_1] - \frac{1}{2}\mathbf{p}_1 \cdot \boldsymbol{\chi}\mathbf{p}_1 \mathrm{Tr}\mathbf{F}_1,$$

and hence the electrostrictive tensor $\mathbb{A}$ defined by (20) satisfies

$$\mathbb{A}(\nabla\xi \otimes \nabla\xi) = \frac{1}{2}[\nabla\xi \otimes (\boldsymbol{\epsilon}\nabla\xi) + (\boldsymbol{\epsilon}\nabla\xi) \otimes \nabla\xi - \mathbf{I}(\nabla\xi \cdot \boldsymbol{\epsilon}\nabla\xi)]. \tag{25}$$

If the material is isotropic with permittivity tensor $\boldsymbol{\epsilon} = \epsilon\mathbf{I}$, we have

$$(\mathbb{A})_{ijkl} = \frac{\varepsilon}{2}\mathbb{T}_{ijkl}, \qquad \text{i.e.,} \qquad \mathbb{A}(\nabla\xi \otimes \nabla\xi) = \epsilon\nabla\xi \otimes \nabla\xi - \frac{\epsilon}{2}|\nabla\xi|^2\mathbf{I}. \tag{26}$$

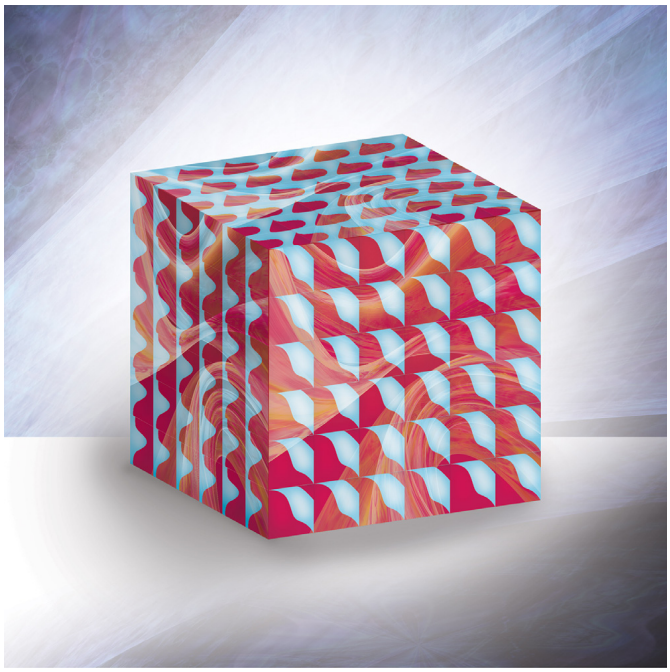

Fig. 3. A schematic of a periodic microstructure. The color in each unit cell could be, in case of electrostrictive composites, heterogeneities, or (in the case of electrets) represent external, immobile positive and negative charges.

Remark 3. In contrast to the $\mathbb{M}$-tensor, the tensor $\mathbb{A}$ defined by (19) and (20) in general does enjoy the minor symmetry, i.e., $(\mathbb{A})_{ijkl} = (\mathbb{A})_{jikl}$, though the Maxwell stress $\boldsymbol{\sigma}_{\text{MW}}$ itself does not have to be symmetric. For example, if the electric susceptibility tensor $\boldsymbol{\chi}$ is anisotropic, the Maxwell stress $\boldsymbol{\sigma}_{\text{MW}}$ defined by (14) is nonsymmetric but the electrostrictive stress (25) is symmetric.

Our interest in this work lies in the electrets, i.e., dielectric soft materials with external immobile charges and dipoles $(\mathbf{p}^e, \rho^e) : D \to \mathbb{R}^3 \times \mathbb{R}$ attached to material points. We assume that the presence of these external charges and dipoles do not alter the material constitutive properties in the sense that the internal energy density function Ψ remains the same as (4), and hence the variational principle (6) for the equilibrium state remains valid. However, the presence of external charges and dipoles does alter the electric field since, by the Maxwell equation, Eq. (3) should be revised to

$$\nabla \cdot [-\epsilon_0 J\mathbf{C}^{-1}\nabla\xi + \mathbf{F}^{-1}(\mathbf{p} + \mathbf{p}^e)] = \rho^e \quad \text{in } D. \tag{27}$$

Again, in the regime of small deformation and moderately small electric field as prescribed by (11), and hence the counterpart of $(19)_1$ can be written as

$$\nabla \cdot (-\boldsymbol{\epsilon}\nabla\xi + \mathbf{p}^e) = \rho^e \quad \text{in } D. \tag{28}$$

Our key physical observation is that the external charges and dipoles $(\rho^e, \mathbf{p}^e)$ break the symmetry, change the qualitative behavior of electrets, and give rise to effective piezoelectricity even in *isotropic* materials. We will elaborate on this in due course.

3. Effective properties of electrostrictive composites: A variational definition

The homogenization of electrets, which will be discussed in the next section, requires the consideration of electrets and electrostriction. Accordingly, it is instructive first to establish the effective properties of purely electrostrictive composites assuming that external charges and dipoles are absent. In particular, we emphasize here a variational definition which offers several advantages in terms of establishing bounds and approximations. Several important references exist on this topic (Lefèvre and Lopez-Pamies, 2017; Castañeda, 2001; Smith et al., 2015; Tian, 2007; Tian et al., 2012) and the reader may access much of the literature through these papers and references therein.

Consider an electrostrictive composite with periodic microstructure as shown in Fig. 3. Let $D \subset \mathbb{R}^3$ be the open bounded domain occupied by the composite body and denote the local electric permittivity tensor, stiffness tensor, and electrostrictive coupling tensor by $(\boldsymbol{\epsilon}^{(\delta)}(\mathbf{x}), \mathbb{C}^{(\delta)}(\mathbf{x}), \mathbb{A}^{(\delta)}(\mathbf{x}))$. Let $Y = (0,1)^3 \subset \mathbb{R}^3$ be the rescaled unit cell (or RVE) of the composite. By periodic microstructure we mean that

$$\left(\boldsymbol{\epsilon}^{(\delta)}(\mathbf{x}), \mathbb{C}^{(\delta)}(\mathbf{x}), \mathbb{A}^{(\delta)}(\mathbf{x})\right) = \left(\boldsymbol{\epsilon}_{\#}\left(\frac{\mathbf{x}}{\delta}\right), \mathbb{C}_{\#}\left(\frac{\mathbf{x}}{\delta}\right), \mathbb{A}_{\#}\left(\frac{\mathbf{x}}{\delta}\right)\right), \tag{29}$$

where $\tilde{\mathbf{x}} \mapsto \left(\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}}), \mathbb{C}_{\#}(\tilde{\mathbf{x}}), \mathbb{A}_{\#}(\tilde{\mathbf{x}})\right)$ are Y-periodic functions.

We denote the effective permittivity tensor by $\boldsymbol{\epsilon}^{\text{eff}}$, the effective stiffness tensor by $\mathbb{C}^{\text{eff}}$, and the effective electrostrictive tensor by $\mathbb{A}^{\text{eff}}$. For the periodic composite specified by (29), Tian (2007) and Tian et al. (2012) have shown that these effective tensors are such that for any $\bar{\mathbf{e}} \in \mathbb{R}^3$ and $\bar{\mathbf{H}} \in \mathbb{R}^{3\times 3}$,

$$\begin{aligned}
&\boldsymbol{\epsilon}^{\text{eff}}\bar{\mathbf{e}} = \fint_Y [\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla \xi_{\bar{\mathbf{e}}})],\\
&\mathbb{C}^{\text{eff}}\bar{\mathbf{H}} = \fint_Y [\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla \mathbf{u}_{\bar{\mathbf{H}}}], \qquad \text{and}\\
&\bar{\mathbf{H}} \cdot \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) = \fint_Y [\nabla \mathbf{u}_{\bar{\mathbf{H}}} \cdot \mathbb{A}_{\#}(\tilde{\mathbf{x}})(\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}})].
\end{aligned} \tag{30}$$

Here, the electric potential $\xi_{\bar{\mathbf{e}}} \in \mathcal{P}_{\bar{\mathbf{e}}}$ (resp. displacement $\mathbf{u}_{\bar{\mathbf{H}}} \in \mathcal{U}_{\bar{\mathbf{H}}}$) satisfy the unit cell problem

$$\text{div}[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla \xi_{\bar{\mathbf{e}}})] = 0 \quad \left(\text{ resp. } \quad \text{div}[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla \mathbf{u}_{\bar{\mathbf{H}}}] = 0\right) \qquad \text{in } Y, \tag{31}$$

and the admissible space $\mathcal{P}_{\bar{\mathbf{e}}}$ (resp. $\mathcal{U}_{\bar{\mathbf{H}}}$) is defined as

$$\begin{aligned}
&\mathcal{P}_{\bar{\mathbf{e}}} \equiv \left\{\xi : -\fint_Y \nabla \xi = \bar{\mathbf{e}} \ \text{ and } \ \nabla \xi \text{ is } Y\text{ -periodic } \right\}\\
&\left(\mathcal{U}_{\bar{\mathbf{H}}} \equiv \left\{\mathbf{u} : \fint_Y \nabla \mathbf{u} = \bar{\mathbf{H}} \ \text{ and } \ \nabla \mathbf{u} \text{ is } Y\text{ -periodic } \right\}\right).
\end{aligned} \tag{32}$$

As is well-known in the classic homogenization theory for linear systems, the effective permittivity tensor $\boldsymbol{\epsilon}^{\text{eff}}$ and the effective stiffness tensor $\mathbb{C}^{\text{eff}}$ can be alternatively defined by the variational principles (Berdichevsky, 2010a, 2010b; Milton, 2002, 2016; Willis, 1981) that for any $\bar{\mathbf{e}} \in \mathbb{R}^3$ and $\bar{\mathbf{H}} \in \mathbb{R}^{3\times 3}$:

$$\begin{cases}
\bar{\mathbf{e}} \cdot \boldsymbol{\epsilon}^{\text{eff}}\bar{\mathbf{e}} = \min\left\{\fint_Y \nabla \xi \cdot \boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla \xi : \ \ \xi \in \mathcal{P}_{\bar{\mathbf{e}}}\right\},\\
\bar{\mathbf{H}} \cdot \mathbb{C}^{\text{eff}}\bar{\mathbf{H}} = \min\left\{\fint_Y \nabla \mathbf{u} \cdot \mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla \mathbf{u} : \ \ \mathbf{u} \in \mathcal{U}_{\bar{\mathbf{H}}}\right\}.
\end{cases} \tag{33}$$

Our first observation is that the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$, together with the effective stiffness tensor $\mathbb{C}^{\text{eff}}$, can be alternatively defined via a variational principle

$$\begin{aligned}
&\Phi_0^{\text{eff}}(\bar{\mathbf{H}}, \bar{\mathbf{e}}) := \min\left\{\fint_Y \nabla \mathbf{u} \cdot \left[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla \mathbf{u} + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}\right] : \ \ \mathbf{u} \in \mathcal{U}_{\bar{\mathbf{H}}}\right\},\\
&\Phi_0^{\text{eff}}(\bar{\mathbf{H}}, \bar{\mathbf{e}}) =: \frac{1}{2}\bar{\mathbf{H}} \cdot \mathbb{C}^{\text{eff}}\bar{\mathbf{H}} + \bar{\mathbf{H}} \cdot \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) - \phi_0(\bar{\mathbf{e}}),
\end{aligned} \tag{34}$$

where $\xi_{\bar{\mathbf{e}}} \in \mathcal{P}_{\bar{\mathbf{e}}}$ is determined by the first of (31) and $\phi_0(\bar{\mathbf{e}})$ will be specified later by (42). In other words, the effective stiffness tensor $\mathbb{C}^{\text{eff}}$ and effective electrostrictive coupling tensor $\mathbb{A}^{\text{eff}}$ can be defined as

$$\mathbb{C}^{\text{eff}} = \left.\frac{\partial^2 \Phi_0^{\text{eff}}}{\partial \bar{\mathbf{H}} \partial \bar{\mathbf{H}}}\right|_{(\bar{\mathbf{H}}, \bar{\mathbf{e}})=0} \qquad \text{and} \qquad \mathbb{A}^{\text{eff}} = \frac{1}{2}\left.\frac{\partial^3 \Phi_0^{\text{eff}}}{\partial \bar{\mathbf{H}} \partial \bar{\mathbf{e}} \partial \bar{\mathbf{e}}}\right|_{(\bar{\mathbf{H}}, \bar{\mathbf{e}})=0}, \tag{35}$$

respectively.

To see the consistency between (34) and (35) and $(30)_{2,3}$, we notice that a minimizer $\mathbf{u}_{\bar{\mathbf{H}}}^* \in \mathcal{U}_{\bar{\mathbf{H}}}$ of the variational problem (34) satisfies

$$\text{div}[\mathbb{C}_{\#}\nabla \mathbf{u}_{\bar{\mathbf{H}}}^* + \mathbb{A}_{\#}\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}] = 0 \quad \text{ in } Y. \tag{36}$$

Let $\mathbf{u}^0 = \mathbf{u}_{\bar{\mathbf{H}}}^* - \mathbf{u}_{\bar{\mathbf{H}}}$. By the second of (31) and (36), we see that $\mathbf{u}^0 \in \mathcal{U}_0$ satisfies

$$\text{div}[\mathbb{C}_{\#}\nabla \mathbf{u}^0 + \mathbb{A}_{\#}\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}] = 0 \quad \text{ in } Y, \tag{37}$$

implying that the displacement $\mathbf{u}^0$ is independent of the average strain $\bar{\mathbf{H}}$. By the divergence theorem, the second of (31) and (37) imply the following identities:

$$\fint_Y \nabla \mathbf{u}^0 \cdot \mathbb{C}_{\#}\nabla \mathbf{u}_{\bar{\mathbf{H}}} = -\fint_Y \mathbf{u}^0 \cdot \text{div}(\mathbb{C}_{\#}\nabla \mathbf{u}_{\bar{\mathbf{H}}}) = 0, \tag{38}$$

$$\fint_Y \nabla \mathbf{u}_{\bar{\mathbf{H}}} \cdot \mathbb{C}_{\#}\nabla \mathbf{u}_{\bar{\mathbf{H}}} = \bar{\mathbf{H}} \cdot \fint_Y \mathbb{C}_{\#}\nabla \mathbf{u}_{\bar{\mathbf{H}}} = \bar{\mathbf{H}} \cdot \mathbb{C}^{\text{eff}}\bar{\mathbf{H}}, \tag{39}$$

and

$$\fint_Y \nabla \mathbf{u}^0 \cdot [\mathbb{C}_{\#}\nabla \mathbf{u}^0 + \mathbb{A}_{\#}\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}] = -\fint_Y \mathbf{u}^0 \cdot \text{div}[\mathbb{C}_{\#}\nabla \mathbf{u}^0 + \mathbb{A}_{\#}\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}] = 0. \tag{40}$$

Therefore,

$$\begin{aligned}\Phi_0^{\text{eff}}(\bar{\mathbf{H}},\bar{\mathbf{e}}) &= \fint_Y \nabla \mathbf{u}^*_{\bar{\mathbf{H}}} \cdot \left[\frac{1}{2}\mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}^*_{\bar{\mathbf{H}}} + \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}\right] \\ &= \fint_Y \nabla(\mathbf{u}^0 + \mathbf{u}_{\bar{\mathbf{H}}}) \cdot \left[\frac{1}{2}\mathbb{C}_\#(\tilde{\mathbf{x}})\nabla(\mathbf{u}^0 + \mathbf{u}_{\bar{\mathbf{H}}}) + \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}\right] \\ &= \fint_Y \Big\{ \frac{1}{2}\nabla \mathbf{u}_{\bar{\mathbf{H}}} \cdot \mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}_{\bar{\mathbf{H}}} + \nabla \mathbf{u}^0 \cdot \mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}_{\bar{\mathbf{H}}} + \nabla \mathbf{u}_{\bar{\mathbf{H}}} \cdot \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}} \\ &\quad + \nabla \mathbf{u}^0 \cdot \left[\frac{1}{2}\mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}^0 + \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}\right]\Big\} \\ &= \frac{1}{2}\bar{\mathbf{H}} \cdot \mathbb{C}^{\text{eff}}\bar{\mathbf{H}} + \bar{\mathbf{H}} \cdot \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) - \phi_0(\bar{\mathbf{e}}), \end{aligned} \tag{41}$$

where the last equality follows from the definitions in (30), (38) and (39). In addition, by (40) we identify

$$\phi_0(\bar{\mathbf{e}}) = -\fint_Y \nabla \mathbf{u}^0 \cdot \left[\frac{1}{2}\mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}^0 + \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}\right] = \frac{1}{2}\fint_Y \nabla \mathbf{u}^0 \cdot \mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}^0. \tag{42}$$

We reiterate that the variational definition (34) and (35) of the effective electrostrictive coupling tensor $\mathbb{A}^{\text{eff}}$ is useful, particularly for obtaining rigorous bounds on the effective properties. Moreover, it implies the interpretation of the effective stiffness tensor $\mathbb{C}^{\text{eff}}$ and the effective electrostrictive coupling tensor $\mathbb{A}^{\text{eff}}$ as the linear mappings such that

$$\mathbb{C}^{\text{eff}}\bar{\mathbf{H}} + \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) = \fint_Y [\mathbb{C}_\#(\tilde{\mathbf{x}})\nabla \mathbf{u}^*_{\bar{\mathbf{H}}} + \mathbb{A}_\#(\tilde{\mathbf{x}})\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}], \tag{43}$$

which may be regarded as a generalization of the classic Hill's relation (Hill, 1963) to electrostrictive composites. To see this, we differentiate (41) with respect to $\bar{\mathbf{H}}$. The last line of (41) yields the left-hand side of (43) whereas the first line of (41) yields the right-hand side of (43).

4. Effective electro-elastic properties of electrets

4.1. Problem formulation

We now consider electrets, i.e., electrostrictive composites with pre-doped external immobile charges and dipoles. Let $(\mathbf{p}^{(\delta)}, \rho^{(\delta)}) : D \to \mathbb{R}^3 \times \mathbb{R}$ be the density of the external dipoles and charges. Here and subsequently, we drop the superscript ^{e} on $(\mathbf{p}^{(\delta)}, \rho^{(\delta)})$ for clarity and tacitly understand that they are "external" source terms for the electric field. Moreover, the scaling parameter δ reflects the fine microstructure of the composite as compared with the macroscopic length-scale of the domain D. We are interested in the macroscopic behavior of the electret in the asymptotic limit $\delta \to 0$.

For simplicity, we consider electrets with periodic distribution of materials (i.e., (29)) and external charges and dipoles (cf., Remark 5):

$$(\mathbf{p}^{(\delta)}, \rho^{(\delta)}) = \chi_D\big(\bar{\mathbf{p}} + \mathbf{p}_\#(\tfrac{\mathbf{x}}{\delta}), \bar{\rho} + \tfrac{1}{\delta}\rho_\#(\tfrac{\mathbf{x}}{\delta})\big), \tag{44}$$

where $(\mathbf{p}_\#, \rho_\#) : \mathbb{R}^3 \to \mathbb{R}^3 \times \mathbb{R}$ are Y-periodic functions , χ_D ($= 1$ on D; $= 0$ otherwise) is the characteristic function of domain D, and

$$\fint_Y (\mathbf{p}_\#, \rho_\#) = 0.$$

We remark that the scaling (44) represents the particular physical scenario that the density and magnitude of external charges and dipoles are invariant for any subdomain $D_1 \subset D$ as $\delta \to 0$:

$$\fint_{D_1} \big(\mathbf{p}^{(\delta)}, \rho^{(\delta)}\big) \to (\overline{\mathbf{p}}, \overline{\rho}). \tag{45}$$

Also, all multipoles beyond the dipole induced by $(\mathbf{p}^{(\delta)}, \rho^{(\delta)})$ vanish as $\delta \to 0$ ($|\alpha| = \alpha_1 + \alpha_2 + \alpha_3$):

$$\begin{aligned} &\fint_{D_1} x_1^{\alpha_1} x_2^{\alpha_2} x_3^{\alpha_3} \mathbf{p}^{(\delta)} \to 0 \quad \text{if } |\alpha| \geq 1, \\ &\fint_{D_1} x_1^{\alpha_1} x_2^{\alpha_2} x_3^{\alpha_3} \rho^{(\delta)} \to 0 \quad \text{if } |\alpha| \geq 2. \end{aligned} \tag{46}$$

Because of the particular scaling (44) and properties (45) and (46), the asymptotic fields and energy can be conveniently calculated by the method of two-scale convergence. There are, however, physical situations where the scalings (44) may not be appropriate (James and Muller, 1994), requiring more careful considerations.

Without loss of generality, we consider the Dirichlet-type boundary conditions (1) for the electrostatic potential and displacement on ∂D. In the regime of "small strain and moderately small electric field", the local electric field is uncoupled

with the elasticity and determined by

$$\begin{cases} \mathrm{div}[-\boldsymbol{\epsilon}^{(\delta)}\nabla\xi^{(\delta)} + \mathbf{p}^{(\delta)}] = \rho^{(\delta)} & \text{in } D, \\ \xi^{(\delta)} = \xi_b & \text{on } \partial D. \end{cases} \tag{47}$$

The electric field induces a Maxwell stress given by $\mathbb{A}^{(\delta)}\nabla\xi^{(\delta)} \otimes \nabla\xi^{(\delta)}$ and henceforth, the mechanical equilibrium equations take the form of

$$\begin{cases} \mathrm{div}[\mathbb{C}^{(\delta)}\nabla\mathbf{u}^{(\delta)} + \mathbb{A}^{(\delta)}\nabla\xi^{(\delta)} \otimes \nabla\xi^{(\delta)}] = 0 & \text{in } D, \\ \mathbf{u}^{(\delta)} = \mathbf{u}_b & \text{on } \partial D. \end{cases} \tag{48}$$

In the next three sections, we proceed to the multiscale analysis of (47) and (48) based on the method of two-scale convergence (Cioranescu and Donato, 1999; Milton, 2002).

Remark 4. From the viewpoint of the electrostatic problem (47), a distribution of dipoles $\mathbf{p}^{(\delta)} : D \to \mathbb{R}^3$ is equivalent to a distribution of charges $\rho'^{(\delta)} = -\nabla\cdot(\mathbf{p}^{(\delta)}\chi_D)$. Conversely, for a given charge distribution $\rho^{(\delta)} : D \to \mathbb{R}^3$ we can solve for a dipole distribution $\mathbf{p}'^{(\delta)} : D \to \mathbb{R}^3$ such that

$$-\nabla\cdot(\mathbf{p}'^{(\delta)}\chi_D) = \rho^{(\delta)} - \bar{\rho} \qquad \text{in } D. \tag{49}$$

where $\bar{\rho} = \fint_D \rho^{(\delta)}$ is the average charge distribution. Therefore, if $\bar{\rho} = 0$, there should be no difference in the electric field and associated mechanical effect induced by $\rho'^{(\delta)}$ and $\mathbf{p}^{(\delta)}$ or by $\rho^{(\delta)}$ and $\mathbf{p}'^{(\delta)}$.

4.2. Multiscale analysis: Electrostatics

We first focus on the electrostatic problem (47). Our goal is to understand the asymptotic limit of the solution $\xi^{(\delta)}$ to (47) as $\delta \to 0$. According to the formal procedure of multiscale analysis, we introduce the 'fast variables' $\tilde{\mathbf{x}} = \mathbf{x}/\delta$ and write the solution $\xi^{(\delta)}$ to (47) as

$$\xi^{(\delta)}(\mathbf{x}) = \xi^{(0)}(\mathbf{x}, \tilde{\mathbf{x}}) + \delta\xi^{(1)}(\mathbf{x}, \tilde{\mathbf{x}}) + \cdots, \tag{50}$$

where $\tilde{\mathbf{x}} \mapsto \xi^{(i)}(\mathbf{x}, \tilde{\mathbf{x}})$ is assumed to be Y-periodic for all i and $\fint_Y \xi^{(i)} = 0$ if $i \neq 0$. Also, from the definition and chain rule, we have the relations:

$$\nabla \to \nabla_{\mathbf{x}} + \frac{1}{\delta}\nabla_{\tilde{\mathbf{x}}}, \qquad \mathrm{div} \to \mathrm{div}_{\mathbf{x}} + \frac{1}{\delta}\mathrm{div}_{\tilde{\mathbf{x}}}. \tag{51}$$

Then the original problem (47) implies the following. First, the leading term is of $O(\frac{1}{\delta^2})$:

$$\frac{1}{\delta^2}\mathrm{div}_{\tilde{\mathbf{x}}}\big[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi^{(0)}\big] = 0. \tag{52}$$

Since $\tilde{\mathbf{x}} \mapsto \xi^{(0)}(\mathbf{x}, \tilde{\mathbf{x}})$ is Y-periodic, a solution to the above equation has to be independent of the fast variable $\tilde{\mathbf{x}}$:

$$\xi^{(0)} = \xi^{(0)}(\mathbf{x}).$$

The next term is of $O(\frac{1}{\delta})$:

$$\tfrac{1}{\delta}\mathrm{div}_{\tilde{\mathbf{x}}}\big\{-\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})[\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}] + \mathbf{p}_{\#}(\tilde{\mathbf{x}})\big\} = \tfrac{1}{\delta}\rho_{\#}(\tilde{\mathbf{x}}) \qquad \forall\ (\mathbf{x}, \tilde{\mathbf{x}}) \in D \times Y, \tag{53}$$

which motivates the unit cell problem for $\xi^{(1)}_{\bar{\mathbf{e}}} \in \mathcal{P}_0$ and $\bar{\mathbf{e}} \in \mathbb{R}^3$:

$$\mathrm{div}_{\tilde{\mathbf{x}}}[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}} + \bar{\mathbf{e}}) + \mathbf{p}_{\#}] = \rho_{\#} \ \text{ in } Y. \tag{54}$$

We remark that the unit cell problem (54) determines the local oscillatory electric field $-\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}$ as implied by (53) with $-\nabla_{\mathbf{x}}\xi^{(0)} = \bar{\mathbf{e}}$ at a fixed point $\mathbf{x} \in D$. Further, we introduce

$$\begin{aligned} &\xi' := \xi^{(1)}_{\bar{\mathbf{e}}} - \bar{\mathbf{e}}\cdot\tilde{\mathbf{x}} - \xi_{\bar{\mathbf{e}}}, \quad \text{i.e.,} \\ &\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}} = -\nabla_{\tilde{\mathbf{x}}}(\xi_{\bar{\mathbf{e}}} + \xi'), \end{aligned} \tag{55}$$

where $\xi_{\bar{\mathbf{e}}} \in \mathcal{P}_{\bar{\mathbf{e}}}$ is a solution to the first of (31). The difference between (54) and (31) implies that $\xi' \in \mathcal{P}_0$ satisfies

$$\mathrm{div}_{\tilde{\mathbf{x}}}[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi') + \mathbf{p}_{\#}] = \rho_{\#} \qquad \text{in } Y. \tag{56}$$

It is clear that a solution to (56) is independent of the average electric field $\bar{\mathbf{e}}$ which. We by

$$\bar{\mathbf{d}}' = \fint_Y [\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi') + \mathbf{p}_{\#}] = \fint_Y \boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi'). \tag{57}$$

Then by $(30)_1$, (55), and (57), we have

$$\fint_Y \big[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}) + \mathbf{p}_{\#}\big] = \fint_Y \big\{\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})[-\nabla_{\tilde{\mathbf{x}}}(\xi_{\bar{\mathbf{e}}} + \xi')] + \mathbf{p}_{\#}(\tilde{\mathbf{x}})\big\} = \boldsymbol{\epsilon}^{\mathrm{eff}}\bar{\mathbf{e}} + \bar{\mathbf{d}}', \tag{58}$$

where $\boldsymbol{\epsilon}^{\text{eff}}$ is the effective permittivity tensor defined by $(30)_1$. Finally, the $O(1)$-terms in (47) yield that $\forall\ (\mathbf{x}, \tilde{\mathbf{x}}) \in D \times Y$,

$$\operatorname{div}_{\mathbf{x}}\big[-\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})[\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}] + \bar{\mathbf{p}} + \mathbf{p}_{\#}(\tilde{\mathbf{x}})\big] + \operatorname{div}_{\tilde{\mathbf{x}}}\big[-\epsilon_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi^{(2)}\big] = \bar{\rho} + \rho_{\#}(\tilde{\mathbf{x}}). \tag{59}$$

Integrating the above equation over the unit cell Y, by (58) and $(47)_2$ we obtain the boundary value problem for the macroscopic electric potential $\xi^{(0)}$:

$$\begin{cases} \operatorname{div}_{\mathbf{x}}\big(\boldsymbol{\epsilon}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{(0)} + (\bar{\mathbf{p}} + \bar{\mathbf{d}}')\chi_D\big) = \bar{\rho} & \text{in } D, \\ \xi^{(0)} = \xi_b & \text{on } \partial D. \end{cases} \tag{60}$$

Remark 5. For a given composite in applications with RVE of length scale δ_0 ($0 \neq \delta_0 \ll 1$ is fixed), according to the assumption (44) the external charge and dipole distributions can be written as

$$(\rho^{(\delta_0)}(\mathbf{x}), \mathbf{p}^{(\delta_0)}(\mathbf{x})) = \Big(\bar{\rho} + \frac{1}{\delta_0}\rho_{\#}\Big(\frac{\mathbf{x}}{\delta_0}\Big),\ \bar{\mathbf{p}} + \mathbf{p}_{\#}\Big(\frac{\mathbf{x}}{\delta_0}\Big)\Big), \qquad \fint_Y \rho_{\#}(\tilde{\mathbf{x}}) = 0. \tag{61}$$

From the viewpoint of fundamental physics and taking cognizance of Remark 4, the microscopic oscillatory electric field $-\nabla_{\tilde{\mathbf{x}}}\xi'$ as determined by (56) can be attributed to an oscillatory charge distribution alone:

$$\operatorname{div}_{\tilde{\mathbf{x}}}[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi')] = \tilde{\rho}_{\#}(\tilde{\mathbf{x}}) := \rho_{\#}(\tilde{\mathbf{x}}) - \nabla_{\tilde{\mathbf{x}}} \cdot \mathbf{p}_{\#}(\tilde{\mathbf{x}}).$$

Conversely, if $\mathbf{p}'_{\#}$ satisfies that $-\nabla_{\tilde{\mathbf{x}}} \cdot (\mathbf{p}'_{\#}(\tilde{\mathbf{x}})) = \rho_{\#}(\tilde{\mathbf{x}})$ in Y, we can alternatively attribute the microscopic oscillatory electric field to an oscillatory dipole distribution alone

$$\operatorname{div}_{\tilde{\mathbf{x}}}[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(-\nabla_{\tilde{\mathbf{x}}}\xi') + \tilde{\mathbf{p}}_{\#}(\tilde{\mathbf{x}})] = 0,$$

where $\tilde{\mathbf{p}}_{\#}(\tilde{\mathbf{x}}) = \mathbf{p}_{\#}(\tilde{\mathbf{x}}) + \mathbf{p}'_{\#}(\tilde{\mathbf{x}})$.

4.3. Multiscale analysis: Elasticity

For the elasticity problem (48), in analogy with the analysis of electrostatic problem (47) we write the solution as

$$\mathbf{u}^{(\delta)}(\mathbf{x}) = \mathbf{u}^{(0)}(\mathbf{x}, \tilde{\mathbf{x}}) + \delta\mathbf{u}^{(1)}(\mathbf{x}, \tilde{\mathbf{x}}) + \cdots, \tag{62}$$

where we recall that $\tilde{\mathbf{x}} = \mathbf{x}/\delta$ is the 'fast' variable and $\mathbf{u}^{(n)}(\mathbf{x}, \tilde{\mathbf{x}})$ are periodic functions of $\tilde{\mathbf{x}}$ with period Y and $\fint_Y \mathbf{u}^{(n)} = 0$ if $n \neq 0$. Inserting (62) into $(48)_1$, by (51) we obtain the leading $O(\frac{1}{\delta^2})$-terms:

$$\frac{1}{\delta^2}\operatorname{div}_{\tilde{\mathbf{x}}}[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(0)}(\mathbf{x}, \tilde{\mathbf{x}})] = 0 \qquad \forall\ (\mathbf{x}, \tilde{\mathbf{x}}) \in D \times Y.$$

Being periodic in $\tilde{\mathbf{x}}$, a solution to the above equation has to be independent of $\tilde{\mathbf{x}}$:

$$\mathbf{u}^{(0)} = \mathbf{u}^{(0)}(\mathbf{x}).$$

Next we collect all the $O(1/\delta)$-terms:

$$0 = \tfrac{1}{\delta}\operatorname{div}_{\tilde{\mathbf{x}}}\Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})[\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}(\mathbf{x}, \tilde{\mathbf{x}}) + \nabla_{\mathbf{x}}\mathbf{u}^{(0)}(\mathbf{x})] + \mathbb{A}_{\#}(\tilde{\mathbf{x}})(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}) \otimes (\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)})\Big\}. \tag{63}$$

In analogy with (53), the above equation motivates the following unit cell problem for $\mathbf{u}^{(1)}_{\bar{\mathbf{H}}} \in \mathcal{U}_0$:

$$\operatorname{div}_{\tilde{\mathbf{x}}}\Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})(\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}_{\bar{\mathbf{H}}} + \bar{\mathbf{H}}) + \mathbb{A}_{\#}(\tilde{\mathbf{x}})(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}) \otimes (\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}})\Big\} = 0. \tag{64}$$

where the average strain $\bar{\mathbf{H}} \in \mathbb{R}^{3\times3}$ in the unit cell Y can be arbitrarily prescribed. Recall that $\mathbf{u}^*_{\bar{\mathbf{H}}}$ is defined by (36). Parallel to (55) we introduce

$$\begin{aligned} &\mathbf{u}' = \mathbf{u}^{(1)}_{\bar{\mathbf{H}}} + \bar{\mathbf{H}}\tilde{\mathbf{x}} - \mathbf{u}^*_{\bar{\mathbf{H}}}, \qquad \text{i.e.,} \\ &\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}_{\bar{\mathbf{H}}} + \bar{\mathbf{H}} = \nabla_{\tilde{\mathbf{x}}}(\mathbf{u}^*_{\bar{\mathbf{H}}} + \mathbf{u}'), \end{aligned} \tag{65}$$

and by (55), rewrite (64) as

$$\operatorname{div}_{\tilde{\mathbf{x}}}\Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})[\nabla_{\tilde{\mathbf{x}}}(\mathbf{u}^*_{\bar{\mathbf{H}}} + \mathbf{u}')] + \mathbb{A}_{\#}(\tilde{\mathbf{x}})[(\nabla_{\tilde{\mathbf{x}}}\xi' + \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}) \otimes (\nabla_{\tilde{\mathbf{x}}}\xi' + \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}})]\Big\} = 0. \tag{66}$$

By (36) we obtain

$$\operatorname{div}_{\tilde{\mathbf{x}}}\Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}' + \mathbb{A}_{\#}(\tilde{\mathbf{x}})[\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi' + 2\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}]\Big\} = 0. \tag{67}$$

Clearly, a solution $\mathbf{u}' \in \mathcal{U}_0$ to the above equation is independent of the average strain $\bar{\mathbf{H}}$.

For convenience in what will follow, we decompose the solution to (67) into two parts, $\mathbf{u}' = \mathbf{u}'_1 + \mathbf{u}'_2$, so that $\mathbf{u}'_1$ depends on $\bar{\mathbf{e}}$ (linearly) while $\mathbf{u}'_2$ is independent of $\bar{\mathbf{e}}$. That is

$$\begin{cases} \operatorname{div}_{\tilde{\mathbf{x}}}\big[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'_1 + 2\mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\big)\big] = 0, \\ \operatorname{div}_{\tilde{\mathbf{x}}}\big[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'_2 + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi'\big)\big] = 0. \end{cases} \tag{68}$$

Solutions to the above linear differential equations enable us to introduce a third-order tensor $\mathbb{B}^{\text{eff}}$ and a second order tensor $\boldsymbol{\sigma}^0$ as

$$\mathbb{B}^{\text{eff}}\bar{\mathbf{e}} = \fint_Y \big[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'_1 + 2\mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\big)\big] \tag{69}$$

and

$$\boldsymbol{\sigma}^0 = \fint_Y \big[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'_2 + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\tilde{\mathbf{x}}}\xi' \otimes \nabla_{\tilde{\mathbf{x}}}\xi'\big)\big], \tag{70}$$

respectively. From the above definitions, it is clear that ($\mathbb{B}^{\text{eff}}$, $\boldsymbol{\sigma}^0$) are independent of the average electric field and strain ($\bar{\mathbf{e}}, \bar{\mathbf{H}}$).

We now calculate the average stress in the unit cell

$$\begin{aligned} &\fint_Y \Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})\big[\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}_{\bar{\mathbf{H}}}(\tilde{\mathbf{x}}) + \bar{\mathbf{H}}\big] + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\big) \otimes \big(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\big)\Big\} \\ &= \fint_Y \Big\{\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^*_{\bar{\mathbf{H}}} + \mathbb{A}_{\#}(\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}} \otimes \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}) + \mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}' + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\big[\nabla\xi' \otimes \nabla\xi' + 2\nabla\xi' \otimes \nabla\xi_{\bar{\mathbf{e}}}\big]\Big\} \\ &= \mathbb{C}^{\text{eff}}\bar{\mathbf{H}} + \mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) + \mathbb{B}^{\text{eff}}\bar{\mathbf{e}} + \boldsymbol{\sigma}^0, \end{aligned} \tag{71}$$

where the first equality follows from inserting (55) and (65), and the last equality follows from (43) and (69). Finally, the $O(1)$-terms of $(48)_1$ implies that $\forall\ (\mathbf{x}, \tilde{\mathbf{x}}) \in D \times Y$,

$$\begin{aligned} \operatorname{div}_{\mathbf{x}}\Big\{&\mathbb{C}_{\#}(\tilde{\mathbf{x}})\big[\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}(\mathbf{x}, \tilde{\mathbf{x}}) + \nabla_{\mathbf{x}}\mathbf{u}^{(0)}(\mathbf{x})\big] \\ &+ \mathbb{A}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\big) \otimes \big(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\big)\Big\} = 0. \end{aligned}$$

Upon integrating over Y, by (71) and $(48)_2$ we obtain the boundary value problem for the macroscopic displacement $\mathbf{u}^{(0)}$:

$$\begin{cases} \operatorname{div}_{\mathbf{x}}\boldsymbol{\sigma} = 0, & \text{in } D, \\ \mathbf{u}^{(0)} = \mathbf{u}_b & \text{on } \partial D, \end{cases} \tag{72}$$

where

$$\boldsymbol{\sigma} \equiv \mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{(0)} - \mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{(0)} + \mathbb{A}^{\text{eff}}\big(\nabla_{\mathbf{x}}\xi^{(0)} \otimes \nabla_{\mathbf{x}}\xi^{(0)}\big) + \boldsymbol{\sigma}^0$$

can be interpreted as the *total* stress in the electret.

Remark 6. From the definition (69), we see that heterogeneity in either elastic properties or dielectric properties is necessary for a nontrivial effective piezoelectricity tensor $\mathbb{B}^{\text{eff}}$. However, the experimental measurements of piezoelectricity of electrets also include another contribution which, termed "apparent piezoelectricity", does not require elastic or dielectric heterogeneity. The distinction between these will be carefully discussed in Section 5.

4.4. Multiscale analysis: Variational approach

It is instructive to carry out the multiscale analyses in Sections 4.2 and 4.3 from a variational perspective. Besides being useful for rigorous bounds, the variational approach yields relations for effective properties that are convenient for extending the present definitions of effective properties to include elastic and electrostatic effects of interfaces.

We first consider the electrostatic problem (47) and introduce the classical Dirichlet energy functional

$$I^{(\delta)}[\xi] = \int_D \Big[\frac{1}{2}\nabla\xi \cdot \boldsymbol{\epsilon}^{(\delta)}\nabla\xi - \mathbf{p}^{(\delta)} \cdot \nabla\xi + \rho^{(\delta)}\xi\Big] \tag{73}$$

By the standard calculation of first variations, we see that (47) is precisely the Euler–Lagrange equation associated with the variational principle

$$\min_{\xi \in \mathcal{H}} I^{(\delta)}[\xi], \qquad \mathcal{H} := \Big\{\xi : D \to \mathbb{R} \;\Big|\; \int_D |\nabla\xi|^2 < +\infty, \xi = \xi_b \text{ on } \partial D\Big\}. \tag{74}$$

In account of the fine microstructure of the composite, we restrict ourselves to test solutions $\xi^{(\delta)}$ that admit the multiscale expansion (50) and recall the identity that for any Y-periodic function $\tilde{\mathbf{x}} \mapsto f(\mathbf{x}, \tilde{\mathbf{x}})$,

$$\int_D f\Big(\mathbf{x}, \frac{\mathbf{x}}{\delta}\Big)d\mathbf{x} = \int_D \fint_Y f(\mathbf{x}, \tilde{\mathbf{x}})d\tilde{\mathbf{x}}d\mathbf{x} + o(1). \tag{75}$$

Inserting (50) into (73), by (75) we have

$$I^{(\delta)}\big[\xi^{(\delta)}\big] = \frac{1}{2\delta^2}\int_D \fint_Y \nabla_{\tilde{\mathbf{x}}}\xi^{(0)}\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi^{(0)} + \frac{1}{\delta}\int_D \fint_Y \Big[\nabla_{\tilde{\mathbf{x}}}\xi^{(0)}\cdot\big(2\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\mathbf{x}}\xi^{(0)} - \mathbf{p}_{\#}\big) + \xi^{(0)}\rho_{\#}\Big] + I^{2S}\big[\xi^{(0)},\xi^{(1)}\big] + o(1), \tag{76}$$

where

$$I^{2S}\big[\xi^{(0)},\xi^{(1)}\big] := \int_D \Big[\fint_Y \frac{1}{2}\big(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\big)\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\big(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\big) - \mathbf{p}_{\#}(\tilde{\mathbf{x}})\cdot\big(\nabla_{\mathbf{x}}\xi^{(0)} + \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\big) - \bar{\mathbf{p}}\cdot\nabla_{\mathbf{x}}\xi^{(0)} + \rho_{\#}\xi^{(1)} + \bar{\rho}\xi^{(0)}\Big]. \tag{77}$$

To keep the functional (76) finite as $\delta\to 0$, we infer that $\fint_Y|\nabla_{\tilde{\mathbf{x}}}\xi^{(0)}|^2 = 0$, i.e., $\xi^{(0)} = \xi^{(0)}(\mathbf{x},\tilde{\mathbf{x}})$ must be independent of the fast variable $\tilde{\mathbf{x}}$. Neglecting the higher-order term $o(1)$ in (76), the variational problem (74) can be recast as

$$\min_{\xi^{(0)}\in\mathcal{H}} \min\big\{I^{2S}\big[\xi^{(0)},\xi^{(1)}\big] : \xi^{(1)}(\mathbf{x},\tilde{\mathbf{x}}) \text{ is } Y\text{ -periodic in } \tilde{\mathbf{x}}\big\}. \tag{78}$$

The inner minimization problem in (78) against microscopic oscillatory electric potential $\xi^{(1)}(\mathbf{x},\tilde{\mathbf{x}})$ motivates the following definition of unit cell energy functional:

$$Q^{\text{eff}}(\bar{\mathbf{e}}) := \min_{\xi\in\mathcal{P}_0}\fint_Y\Big[\frac{1}{2}(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi)\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi) + \mathbf{p}_{\#}(\tilde{\mathbf{x}})\cdot(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi) + \rho_{\#}\xi\Big]. \tag{79}$$

It is not hard to see that a solution to the variational problem (79), denoted by $\xi^{(1)}_{\bar{\mathbf{e}}}$, will satisfy the associated Euler–Lagrange equation, i.e., the unit cell problem (54). Therefore,

$$\begin{aligned} Q^{\text{eff}}(\bar{\mathbf{e}}) &= \fint_Y\Big[\frac{1}{2}\big(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\big)\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\big(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\big) + \mathbf{p}_{\#}(\tilde{\mathbf{x}})\cdot\big(\bar{\mathbf{e}} - \nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\big)\Big] \\ &= \fint_Y\Big[\frac{1}{2}\nabla_{\tilde{\mathbf{x}}}\big(\xi_{\bar{\mathbf{e}}} + \xi'\big)\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\big(\xi_{\bar{\mathbf{e}}} + \xi'\big) - \mathbf{p}_{\#}(\tilde{\mathbf{x}})\cdot\nabla_{\tilde{\mathbf{x}}}\big(\xi_{\bar{\mathbf{e}}} + \xi'\big)\Big] \\ &= \fint_Y\Big\{\frac{1}{2}\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}} + \nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\cdot\big[\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi' - \mathbf{p}_{\#}(\tilde{\mathbf{x}})\big]\Big\} + Q_0 \\ &= \frac{1}{2}\bar{\mathbf{e}}\cdot\boldsymbol{\epsilon}^{\text{eff}}\bar{\mathbf{e}} + \bar{\mathbf{e}}\cdot\bar{\mathbf{d}}' + Q_0, \end{aligned} \tag{80}$$

where the second equality follows from (55), and the last equality follows from $(33)_1$, (57) and the following definition of constant Q_0:

$$Q_0 := \fint_Y\Big[\frac{1}{2}\nabla_{\tilde{\mathbf{x}}}\xi'\cdot\boldsymbol{\epsilon}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi' - \mathbf{p}_{\#}(\tilde{\mathbf{x}})\cdot\nabla_{\tilde{\mathbf{x}}}\xi' + \rho_{\#}\xi'\Big].$$

Comparing the integrands in (79) and (77), we see that

$$\begin{aligned} &\min_{\xi^{(0)}\in\mathcal{H}} \min\big\{I^{2S}\big[\xi^{(0)},\xi^{(1)}\big] : \xi^{(1)}(\mathbf{x},\tilde{\mathbf{x}}) \text{ is } Y\text{-periodic in } \tilde{\mathbf{x}}\big\} \\ &= \min_{\xi^{(0)}\in\mathcal{H}}\int_D\Big[Q^{\text{eff}}\big(\nabla\xi^{(0)}\big) - \bar{\mathbf{p}}\cdot\nabla_{\mathbf{x}}\xi^{(0)} + \bar{\rho}\xi^{(0)}\Big] \\ &= \min_{\xi^{(0)}\in\mathcal{H}}\Big\{\int_D\Big[\frac{1}{2}\nabla_{\mathbf{x}}\xi^{(0)}\cdot\boldsymbol{\epsilon}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{(0)} - \big(\mathbf{d}' + \bar{\mathbf{p}}\big)\cdot\nabla_{\mathbf{x}}\xi^{(0)} + \bar{\rho}\xi^{(0)}\Big]d\mathbf{x} + const.\Big\}, \end{aligned}$$

from which we observe that the macroscopic electric field $-\nabla_{\mathbf{x}}\xi^{(0)}$ satisfies the boundary value problem (60).

Similarly, for the elastic problem we consider the "linearized" energy functional for the composite body D:

$$F^{(\delta)}[\mathbf{u}] = \int_D\Big[\frac{1}{2}\nabla\mathbf{u}\cdot\mathbb{C}^{(\delta)}\nabla\mathbf{u} + \nabla\mathbf{u}\cdot\mathbb{A}^{(\delta)}\big(\nabla\xi^{(\delta)}\otimes\nabla\xi^{(\delta)}\big)\Big], \tag{81}$$

where $\xi^{(\delta)}$ is the solution to the variational problem (74) or the boundary value problem (47). By the standard calculation of first variations, we see that (48) is precisely the Euler–Lagrange equation associated with the variational principle:

$$\min_{\mathbf{u}\in\mathcal{W}} F^{(\delta)}[\mathbf{u}], \qquad \mathcal{W} := \Big\{\mathbf{u} : D\to\mathbb{R}^3 \;\Big|\; \int_D|\nabla\mathbf{u}|^2 < +\infty,\ \mathbf{u} = \mathbf{u}_b \text{ on } \partial D\Big\}. \tag{82}$$

Restricting ourselves to test solutions $\mathbf{u}^{(\delta)}$ that admit multiscale expansion (62), by (81) and (75) we again infer that $\mathbf{u}^{(0)} = \mathbf{u}^{(0)}(\mathbf{x},\tilde{\mathbf{x}})$ must be independent of the fast variable $\tilde{\mathbf{x}}$ to keep $F^{(\delta)}[\mathbf{u}^{(\delta)}]$ finite, and find that

$$F^{(\delta)}\big[\mathbf{u}^{(\delta)}\big] = F^{2S}\big[\mathbf{u}^{(0)},\mathbf{u}^{(1)}\big] + o(1), \tag{83}$$

where

$$F^{2S}[\mathbf{u}^{(0)},\mathbf{u}^{(1)}] := \int_D \fint_Y \left(\nabla_{\mathbf{x}}\mathbf{u}^{(0)}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}\right)\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\left(\nabla_{\mathbf{x}}\mathbf{u}^{(0)}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{(1)}\right) + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\Big[\left(\nabla_{\mathbf{x}}\xi^{(0)}+\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\right)\otimes\left(\nabla_{\mathbf{x}}\xi^{(0)}+\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}\right)\Big]\Big]. \tag{84}$$

Neglecting the higher-order term $o(1)$ in (83), the variational problem (82) can be recast as

$$\min_{\mathbf{u}^{(0)}\in\mathcal{W}}\min\left\{F^{2S}\left[\mathbf{u}^{(0)},\mathbf{u}^{(1)}\right]:\ \mathbf{u}^{(1)}(\mathbf{x},\tilde{\mathbf{x}})\ \text{is } Y\text{-periodic in } \tilde{\mathbf{x}}\right\}. \tag{85}$$

The inner minimization problem in (85) against microscopic oscillatory displacement $\mathbf{u}^{(1)}(\mathbf{x},\tilde{\mathbf{x}})$ motivates the following definition of unit cell energy function:

$$\Phi^{\text{eff}}(\bar{\mathbf{H}};\bar{\mathbf{e}}) := \min_{\mathbf{u}\in\mathcal{U}_0}\fint_Y \left(\bar{\mathbf{H}}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}\right)\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\left(\bar{\mathbf{H}}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}\right) + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\Big[\left(\bar{\mathbf{e}}-\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\right)\otimes\left(\bar{\mathbf{e}}-\nabla_{\tilde{\mathbf{x}}}\xi^{(1)}_{\bar{\mathbf{e}}}\right)\Big]\Big], \tag{86}$$

where $\xi^{(1)}_{\bar{\mathbf{e}}}$ satisfies the unit cell problem (54). A solution to the above variational problem, denoted by $\mathbf{u}^{(1)}_{\bar{\mathbf{H}}}$, satisfies the associated Euler–Lagrange equation, i.e., the unit cell problem (64). Therefore,

$$\begin{aligned}\Phi^{\text{eff}}\left(\bar{\mathbf{H}};\bar{\mathbf{e}}\right) &= \fint_Y \left(\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{*}_{\bar{\mathbf{H}}}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'\right)\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\left(\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{*}_{\bar{\mathbf{H}}}+\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'\right) \\ &\quad + \mathbb{A}_{\#}(\tilde{\mathbf{x}})\Big[\nabla_{\tilde{\mathbf{x}}}\left(\xi_{\bar{\mathbf{e}}}+\xi'\right)\otimes\nabla_{\tilde{\mathbf{x}}}\left(\xi_{\bar{\mathbf{e}}}+\xi'\right)\Big]\Big] \\ &=: T_0+T_1+T_2 = T_0+\Phi^{\text{eff}}_0\left(\bar{\mathbf{H}};\bar{\mathbf{e}}\right)+\bar{\mathbf{H}}\cdot\left(\mathbb{B}^{\text{eff}}\bar{\mathbf{e}}+\boldsymbol{\sigma}_0\right),\end{aligned} \tag{87}$$

where the first equality follows from (55) and (65), and the last equality follows from the following definitions:

$$T_0 = \fint_Y \nabla\mathbf{u}'\cdot\Big[\tfrac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla\mathbf{u}'+\mathbb{A}_{\#}(\tilde{\mathbf{x}})\Big[\nabla_{\tilde{\mathbf{x}}}(\xi_{\bar{\mathbf{e}}}+\xi')\otimes\nabla_{\tilde{\mathbf{x}}}\left(\xi_{\bar{\mathbf{e}}}+\xi'\right)\Big]\Big], \tag{88}$$

$$T_1 = \fint_Y \nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{*}_{\bar{\mathbf{H}}}\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{*}_{\bar{\mathbf{H}}}+\mathbb{A}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\otimes\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\Big] = \Phi^{\text{eff}}_0\left(\bar{\mathbf{H}},\bar{\mathbf{e}}\right)\quad(\text{cf., (41)}), \tag{89}$$

and

$$\begin{aligned}T_2 &= \fint_Y \nabla_{\tilde{\mathbf{x}}}\mathbf{u}^{*}_{\bar{\mathbf{H}}}\cdot\left[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'+\mathbb{A}_{\#}(\tilde{\mathbf{x}})\left(\nabla_{\tilde{\mathbf{x}}}\xi'\otimes\nabla_{\tilde{\mathbf{x}}}\xi'+2\nabla_{\tilde{\mathbf{x}}}\xi'\otimes\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\right)\right] \\ &= \bar{\mathbf{H}}\cdot\fint_Y\left[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}'+\mathbb{A}_{\#}(\tilde{\mathbf{x}})\left(\nabla_{\tilde{\mathbf{x}}}\xi'\otimes\nabla_{\tilde{\mathbf{x}}}\xi'+2\nabla_{\tilde{\mathbf{x}}}\xi'\otimes\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\right)\right] \\ &= \bar{\mathbf{H}}\cdot\left(\mathbb{B}^{\text{eff}}\bar{\mathbf{e}}+\boldsymbol{\sigma}_0\right)\quad(\text{cf., (67)}-(69)).\end{aligned} \tag{90}$$

We note that T_0 is independent of $\bar{\mathbf{H}}$. Comparing the integrands in (86) and (84), we see that

$$\begin{aligned}&\min_{\mathbf{u}^{(0)}\in\mathcal{W}}\min\left\{F^{2S}\left[\mathbf{u}^{(0)},\mathbf{u}^{(1)}\right]:\mathbf{u}^{(1)}(\mathbf{x},\tilde{\mathbf{x}})\ \text{is } Y\text{ -periodic in } \tilde{\mathbf{x}}\right\} = \min_{\mathbf{u}^{(0)}\in\mathcal{W}}\int_D \Phi^{\text{eff}}\left(\nabla\mathbf{u}^{(0)}\right) \\ &= \min_{\mathbf{u}^{(0)}\in\mathcal{W}}\left\{\int_D\left[\nabla_{\mathbf{x}}\mathbf{u}^{(0)}\cdot\left(\frac{1}{2}\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{(0)}+\mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{(0)}\otimes\nabla_{\mathbf{x}}\xi^{(0)}\right)-\mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{(0)}+\boldsymbol{\sigma}_0\right)\right]+C_0'\right\},\end{aligned}$$

from which we immediately find that the macroscopic displacement $\mathbf{u}^{(0)}$ satisfies the boundary value problem (72).

In conclusion, upon solving the unit cell problems (53) and (64) or directly the minimization problem (86), we can determine the "effective" internal energy density $\Phi^{\text{eff}}=\Phi^{\text{eff}}(\bar{\mathbf{H}},\bar{\mathbf{e}})$ as a function of average strain and electric field $(\bar{\mathbf{H}},\bar{\mathbf{e}})$. From (41) and (87), we find that the functional form of the effective energy density has to be of the following form:

$$\Phi^{\text{eff}}(\bar{\mathbf{H}},\bar{\mathbf{e}}) = \frac{1}{2}\bar{\mathbf{H}}\cdot\mathbb{C}^{\text{eff}}\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\mathbb{A}^{\text{eff}}(\bar{\mathbf{e}}\otimes\bar{\mathbf{e}})+\bar{\mathbf{H}}\cdot\mathbb{B}^{\text{eff}}\bar{\mathbf{e}}+\boldsymbol{\sigma}^0\cdot\bar{\mathbf{H}}-\phi_0(\bar{\mathbf{e}})+T_0(\bar{\mathbf{e}}), \tag{91}$$

where $\boldsymbol{\sigma}^0$ is independent of $(\bar{\mathbf{H}},\bar{\mathbf{e}})$. Therefore, the fourth-order effective stiffness tensor $\mathbb{C}^{\text{eff}}$, third order piezoelectric tensor $\mathbb{B}^{\text{eff}}$ and fourth-order effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$ can be alternatively defined as

$$\mathbb{C}^{\text{eff}} = \frac{\partial^2\Phi^{\text{eff}}}{\partial\bar{\mathbf{H}}\partial\bar{\mathbf{H}}}\Big|_{(\bar{\mathbf{H}},\bar{\mathbf{e}})=0},\quad \mathbb{B}^{\text{eff}} = \frac{\partial^2\Phi^{\text{eff}}}{\partial\bar{\mathbf{H}}\partial\bar{\mathbf{e}}}\Big|_{(\bar{\mathbf{H}},\bar{\mathbf{e}})=0},\quad \mathbb{A}^{\text{eff}} = \frac{1}{2}\frac{\partial^3\Phi^{\text{eff}}}{\partial\bar{\mathbf{H}}\partial\bar{\mathbf{e}}\partial\bar{\mathbf{e}}}\Big|_{(\bar{\mathbf{H}},\bar{\mathbf{e}})=0}. \tag{92}$$

5. The distinction between "apparent" and "effective" piezoelectricity of electrets

As outlined in the introductory section, experiments and past theoretical works have pointed out that electrets display an apparent piezoelectric like effect. In this section, based on the analysis in the preceding sections, we carefully extract the piezoelectric properties of electrets and distinguish between "apparent" piezoelectricity—which is not a true bulk property in the thermodynamic sense—from "effective" piezoelectricity (which is).

From the analyses in Section 4.2–4.4, we see that the macroscopic electric field and strain of the electret are determined by (60) and (72), respectively. In particular, from (72) we see that the electret is *effectively* piezoelectric since the term $-\mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{(0)}$ can be interpreted as the stress induced by electric field. We also notice that the doped immobile dipoles and charges $(\mathbf{p}^{(\delta)}, \rho^{(\delta)})$ have no influence on the effective permittivity tensor $\boldsymbol{\epsilon}^{\text{eff}}$, stiffness tensor $\mathbb{C}^{\text{eff}}$ and electrostrictive tensor $\mathbb{A}^{\text{eff}}$. The third-order piezoelectric coupling tensor $\mathbb{B}^{\text{eff}}$ defined by (69) does depend on the microscopic charge density and polarization $(\rho_{\#}, \mathbf{p}_{\#})$ in the unit cell and microstructure, but is independent of the boundary conditions (1) and the average dipole and charge distributions $(\bar{\mathbf{p}}, \bar{\rho})$.

In an experimental setup, there are additional contributions to the *apparent* piezoelectricity due to the electrostrictive effect that depends on the macroscopic boundary conditions and the average charge and dipole distributions. The reason lies in that experiments measure the response of the electret with reference to certain initial state, e.g., the natural state of the electret when the body D is placed in space and free from external mechanical and electrical loading on its boundary:

$$\xi_b = 0 \qquad \text{and} \qquad \boldsymbol{\sigma}\mathbf{n} = 0 \qquad \text{on } \partial D, \tag{93}$$

where $\mathbf{n}$ is the outward unit normal on ∂D. This initial configuration may admit nonzero displacement and electric field with respect to the reference state defined by (9) because of the presence of externally doped charges and dipoles. From the analysis in prior sections, in particular, (60) and (72), we see that the macroscopic electric field (denoted by $-\nabla\xi^{\text{ini}}$) and displacement (denoted by $\mathbf{u}^{\text{ini}}$) in this *initial* configuration are respectively determined by

$$\begin{cases} \operatorname{div}\left[\boldsymbol{\epsilon}^{\text{eff}}\nabla\xi^{\text{ini}} + (\bar{\mathbf{p}} + \bar{\mathbf{d}}')\chi_D\right] = \bar{\rho} & \text{in } D, \\ \xi^{\text{ini}} = \xi_b = 0 & \text{on } \partial D, \end{cases} \tag{94}$$

and

$$\begin{cases} \operatorname{div}\Big[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{ini}} - \mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{\text{ini}} \\ \qquad + \mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{ini}} \otimes \nabla_{\mathbf{x}}\xi^{\text{ini}}\right) + \boldsymbol{\sigma}^0\Big] = 0 \qquad \text{in } D, \\ \Big[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{ini}} - \mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{\text{ini}} \\ \qquad + \mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{ini}} \otimes \nabla_{\mathbf{x}}\xi^{\text{ini}}\right) + \boldsymbol{\sigma}^0\Big]\mathbf{n} = 0 \qquad \text{on } \partial D. \end{cases} \tag{95}$$

Next, we consider an applied voltage such that the apparent average electric field in the body is given by $\bar{\mathbf{e}}^0 \in \mathbb{R}^3$, i.e., the following boundary conditions

$$\xi_b = -\bar{\mathbf{e}}^0 \cdot \mathbf{x} \qquad \text{and} \qquad \boldsymbol{\sigma}\mathbf{n} = 0 \qquad \text{on } \partial D. \tag{96}$$

Again, by (60) and (72) we infer that the macroscopic electric field (denoted by $-\nabla\xi^{\text{fin}}$) and displacement (denoted by $\mathbf{u}^{\text{fin}}$) in this *final* configuration are respectively determined by

$$\begin{cases} \operatorname{div}\left(\boldsymbol{\epsilon}^{\text{eff}}\nabla\xi^{\text{fin}} + (\bar{\mathbf{p}} + \bar{\mathbf{d}}')\chi_D\right) = \bar{\rho} & \text{in } D, \\ \xi^{\text{fin}} = \xi_b = -\bar{\mathbf{e}}^0 \cdot \mathbf{x} & \text{on } \partial D, \end{cases} \tag{97}$$

and

$$\begin{cases} \operatorname{div}\left[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{fin}} - \mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{\text{fin}} + \mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{fin}} \otimes \nabla_{\mathbf{x}}\xi^{\text{fin}}\right) + \boldsymbol{\sigma}^0\right] = 0 & \text{in } D, \\ \left[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{fin}} - \mathbb{B}^{\text{eff}}\nabla_{\mathbf{x}}\xi^{\text{fin}} + \mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{fin}} \otimes \nabla_{\mathbf{x}}\xi^{\text{fin}}\right) + \boldsymbol{\sigma}^0\right]\mathbf{n} = 0 & \text{on } \partial D. \end{cases} \tag{98}$$

Comparing (94) and (95) with (97) and (98) we see that

$$-\nabla\left(\xi^{\text{fin}} - \xi^{\text{ini}}\right) = \bar{\mathbf{e}}^0 \qquad \text{in } D,$$

and that the change of strain $\nabla\mathbf{u}^{\text{chg}} := \nabla(\mathbf{u}^{\text{fin}} - \mathbf{u}^{\text{ini}})$ satisfies the following boundary value problem:

$$\begin{cases} \operatorname{div}\left[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{chg}} + \mathbb{B}^{\text{eff}}\bar{\mathbf{e}}^0 - 2\mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{ini}} \otimes \bar{\mathbf{e}}^0\right) + \mathbb{A}^{\text{eff}}\left(\bar{\mathbf{e}}^0 \otimes \bar{\mathbf{e}}^0\right)\right] = 0 & \text{in } D, \\ \left[\mathbb{C}^{\text{eff}}\nabla_{\mathbf{x}}\mathbf{u}^{\text{chg}} + \mathbb{B}^{\text{eff}}\bar{\mathbf{e}}^0 - 2\mathbb{A}^{\text{eff}}\left(\nabla_{\mathbf{x}}\xi^{\text{ini}} \otimes \bar{\mathbf{e}}^0\right) + \mathbb{A}^{\text{eff}}\left(\bar{\mathbf{e}}^0 \otimes \bar{\mathbf{e}}^0\right)\right]\mathbf{n} = 0 & \text{on } \partial D. \end{cases} \tag{99}$$

From (99) we may identify the quantity

$$\mathbb{B}^{\text{ap}} = \mathbb{B}^{\text{eff}} - 2\mathbb{A}^{\text{eff}}\nabla\xi^{\text{ini}} \tag{100}$$

as the *apparent* piezoelectric tensor of the composite, which in general depends on the position-$\mathbf{x}$ if the macroscopic initial electric field $-\nabla\xi^{\text{ini}}$ is nonuniform on D. We emphasize that the apparent piezoelectricity arising from the pre-existing electric field (i.e., the second term in (100)) is present even in *homogeneous* dielectric electrostrictive materials.

At this point, it is worthwhile to summarize the effects of the doped external immobile charges and dipoles in electrets.

1. In the absence of external charges and polarization (i.e., $\mathbf{p}_{\#} = \rho_{\#} = 0$), the overall composite behaves like an electrostrictive material with effective stiffness and electrostrictive tensors as defined by (92). The determination of the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$ (cf., $(30)_3$ or (35)) requires the solution to the unit cell problems (31) or the variational problem (34), and is independent of the external charges or dipoles.
2. In the presence of external charges and polarization:

$$\left(\mathbf{p}^{(\delta)}, \rho^{(\delta)}\right) = \chi_D\left(\bar{\mathbf{p}} + \mathbf{p}_{\#}\left(\frac{\mathbf{x}}{\delta}\right), \bar{\rho} + \frac{1}{\delta}\rho_{\#}\left(\frac{\mathbf{x}}{\delta}\right)\right),$$

   the microscopic oscillatory electric field as determined by (54) depends on the microscopic oscillatory dipole and charge densities $(\mathbf{p}_{\#}, \rho_{\#})$ but is independent of the average dipole and charge density $(\bar{\mathbf{p}}, \bar{\rho})$. The effective internal energy density (91) and the effective tensors defined in (92) (e.g., $\mathbb{B}^{\text{eff}}$), do depend on $(\mathbf{p}_{\#}, \rho_{\#})$ but are independent of the average $(\bar{\mathbf{p}}, \bar{\rho})$ or the macroscopic boundary conditions, e.g., (1).
3. If the average polarization and charge density $(\bar{\mathbf{p}}, \bar{\rho}) = 0$, for boundary condition (93) the initial electric field $-\nabla\xi^{\text{ini}}$ defined by (94) vanishes, and hence the apparent piezoelectricity coincides with the effective piezoelectricity by (100).
4. If the average external dipole and charge densities $(\bar{\mathbf{p}}, \bar{\rho})$ are nonzero or the initial macroscopic boundary conditions are nontrivial, an important effect lies in that a nonzero macroscopic electric field is present in the initial state (cf., (94)). In this case, the electro-elastic response of the composite with reference to the initial state behaves as a piezoelectric material with an apparent piezoelectric tensor $\mathbb{B}^{\text{ap}}$ given by (100), which may be nonzero even if $\mathbb{B}^{\text{eff}} = 0$. Therefore, one must be careful in interpreting the observed piezoelectricity in the experiments of electrets and differentiating the two contributions in (100).

6. Applications

The results established in the preceding sections may be used to design next-generation electret materials; especially using numerical tools or topology optimization approaches as in the context of conventional piezoelectricity, (cf., Nanthakumar et al., 2016). Below we present some applications that can be arrived at analytically.

6.1. Rigorous variational bounds for effective electrostrictive tensors

As a first application, we derive rigorous bounds for the effective electrostrictive tensors $\mathbb{A}^{\text{eff}}$ based on our variational definition (34). It will be found convenient to make the following definitions:

$$\bar{\mathbb{C}} = \fint_Y \mathbb{C}_{\#}, \qquad \bar{\mathbb{C}}_H = \left[\fint_Y \mathbb{C}_{\#}^{-1}\right]^{-1}$$

the arithmetic and harmonic mean of the elastic property tensor, respectively. It is well-known that the classic Voigt–Reuss bounds hold for the effective elastic tensor $\mathbb{C}^{\text{eff}}$ (cf., (30), Milton, 2002):

$$\bar{\mathbf{H}} \cdot \bar{\mathbb{C}}_H \bar{\mathbf{H}} \le \bar{\mathbf{H}} \cdot \mathbb{C}^{\text{eff}} \bar{\mathbf{H}} \le \bar{\mathbf{H}} \cdot \bar{\mathbb{C}} \bar{\mathbf{H}} \qquad \forall \bar{\mathbf{H}} \in \mathbb{R}^{3\times 3}.$$

Also, since the electrostatic problem $(31)_1$ has to be *a priori* solved to define the variational principle $(34)_1$, we take the microscopic electrostatic field $-\nabla\xi_{\bar{\mathbf{e}}}$, the effective permittivity and elasticity tensors $(\boldsymbol{\epsilon}^{\text{eff}}, \mathbb{C}^{\text{eff}})$, and the nonnegative qudratic function $\phi_0(\bar{\mathbf{e}})$ defined by (42) as given quantities in our subsequent calculations. For brevity, we introduce quantities:

$$\mathbf{P}_{\bar{\mathbf{e}}} = \mathbb{A}_{\#}(\tilde{\mathbf{x}}) \nabla\xi_{\bar{\mathbf{e}}} \otimes \nabla\xi_{\bar{\mathbf{e}}}, \quad \bar{\mathbf{P}}_{\bar{\mathbf{e}}} = \fint_Y \mathbf{P}_{\bar{\mathbf{e}}}, \quad \mathbf{F}_{\bar{\mathbf{e}}} = \fint_Y \mathbb{C}_{\#}^{-1} \mathbf{P}_{\bar{\mathbf{e}}}. \tag{101}$$

In addition, we restrict ourselves to ideal dielectric materials with the electric permittivity tensor $\boldsymbol{\epsilon}$ and the electrostrictive tensor $\mathbb{A}$ defined by (25). From (25) and (30) we find that

$$\begin{aligned}\bar{\mathbf{P}}_{\bar{\mathbf{e}}} &= \frac{1}{2}\fint_Y \left[\nabla\xi_{\bar{\mathbf{e}}} \otimes (\boldsymbol{\epsilon}_{\#}\nabla\xi_{\bar{\mathbf{e}}}) + (\boldsymbol{\epsilon}_{\#}\nabla\xi_{\bar{\mathbf{e}}}) \otimes \nabla\xi_{\bar{\mathbf{e}}} - \mathbf{I}\nabla\xi_{\bar{\mathbf{e}}} \cdot (\boldsymbol{\epsilon}_{\#}\nabla\xi_{\bar{\mathbf{e}}})\right] \\ &= -\frac{\mathbf{I}}{2}\left(\bar{\mathbf{e}} \cdot \boldsymbol{\epsilon}^{\text{eff}} \bar{\mathbf{e}}\right) + \frac{1}{2}\fint_Y \left[\nabla\xi_{\bar{\mathbf{e}}} \otimes (\boldsymbol{\epsilon}_{\#}\nabla\xi_{\bar{\mathbf{e}}}) + (\boldsymbol{\epsilon}_{\#}\nabla\xi_{\bar{\mathbf{e}}}) \otimes \nabla\xi_{\bar{\mathbf{e}}}\right],\end{aligned}$$

and hence $\text{Tr}\bar{\mathbf{P}}_{\bar{\mathbf{e}}} = -\frac{1}{2}(\bar{\mathbf{e}} \cdot \boldsymbol{\epsilon}^{\text{eff}} \bar{\mathbf{e}})$ in $\mathbb{R}^3$.

For a Voigt-type bound on the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$, we choose a particular test strain field $\nabla\mathbf{u} = \bar{\mathbf{H}}$ for the minimization problem $(34)_1$ and obtain that

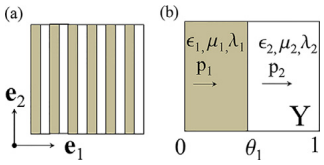

Fig. 4. An electret of two ideal dielectric materials: (a) A simple laminate, (b) a rescaled unit cell.

$$\begin{aligned}\Phi_0^{\text{eff}}(\bar{\mathbf{H}},\bar{\mathbf{e}}) &\le \fint_Y \tilde{\mathbf{H}}\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\tilde{\mathbf{H}}+\mathbb{A}_{\#}(\tilde{\mathbf{x}})(\nabla\xi_{\bar{\mathbf{e}}}\otimes\nabla\xi_{\bar{\mathbf{e}}})\Big]\\ &= \frac{1}{2}\bar{\mathbf{H}}\cdot\bar{\mathbb{C}}\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\fint_Y \mathbb{A}_{\#}(\tilde{\mathbf{x}})(\nabla\xi_{\bar{\mathbf{e}}}\otimes\nabla\xi_{\bar{\mathbf{e}}})\\ &= \frac{1}{2}\bar{\mathbf{H}}\cdot\bar{\mathbb{C}}\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\bar{\mathbf{P}}_{\bar{\mathbf{e}}}.\end{aligned}$$

By (41) we conclude that $\forall\,\bar{\mathbf{H}}\in\mathbb{R}^{3\times3},\ \bar{\mathbf{e}}\in\mathbb{R}^3$,

$$\bar{\mathbf{H}}\cdot\mathbb{A}^{\text{eff}}(\bar{\mathbf{e}}\otimes\bar{\mathbf{e}}) \le \frac{1}{2}\bar{\mathbf{H}}\cdot\big(\bar{\mathbb{C}}-\mathbb{C}^{\text{eff}}\big)\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\bar{\mathbf{P}}_{\bar{\mathbf{e}}}+\phi_0(\bar{\mathbf{e}}), \tag{102}$$

where $\phi_0(\bar{\mathbf{e}})$ is defined by (42).

For a Reuss-type bound, neglecting the differential constraint on the strain in $(34)_1$ we have

$$\Phi_0^{\text{eff}}(\bar{\mathbf{H}},\bar{\mathbf{e}}) \ge \min\Big\{\fint_Y \mathbf{M}(\tilde{\mathbf{x}})\cdot\Big[\frac{1}{2}\mathbb{C}_{\#}(\tilde{\mathbf{x}})\mathbf{M}(\tilde{\mathbf{x}})+\mathbb{A}_{\#}(\tilde{\mathbf{x}})\nabla\xi_{\bar{\mathbf{e}}}\otimes\nabla\xi_{\bar{\mathbf{e}}}\Big]:\ \mathbf{M}\in\mathcal{M}_{\bar{\mathbf{H}}}\Big\}, \tag{103}$$

where the functional space $\mathcal{M}_{\bar{\mathbf{H}}}$ includes all Y-periodic tensor fields with average $\bar{\mathbf{H}}$:

$$\mathcal{M}_{\bar{\mathbf{H}}} := \Big\{\mathbf{M}: Y\to\mathbb{R}^3 \text{ is } Y\text{ -periodic and } \fint_Y \mathbf{M}=\bar{\mathbf{H}}\Big\}.$$

The minimization problem on the right hand side of (103) can be explicitly solved by the method of Lagrange's multiplier, and the minimum is given by

$$\frac{1}{2}\bar{\mathbf{H}}\cdot\bar{\mathbb{C}}_H\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\bar{\mathbb{C}}_H\mathbf{F}_{\bar{\mathbf{e}}}+\frac{1}{2}\Big[\mathbf{F}_{\bar{\mathbf{e}}}\cdot\bar{\mathbb{C}}_H\mathbf{F}_{\bar{\mathbf{e}}}-\fint_Y \mathbf{P}\cdot\mathbb{C}_{\#}^{-1}\mathbf{P}\Big].$$

Inserting $(34)_2$ into (103), we conclude that

$$\begin{aligned}\bar{\mathbf{H}}\cdot\mathbb{A}^{\text{eff}}(\bar{\mathbf{e}}\otimes\bar{\mathbf{e}}) \ge{}& \frac{1}{2}\bar{\mathbf{H}}\cdot\big(\bar{\mathbb{C}}_H-\mathbb{C}^{\text{eff}}\big)\bar{\mathbf{H}}+\bar{\mathbf{H}}\cdot\bar{\mathbb{C}}_H\mathbf{F}_{\bar{\mathbf{e}}}\\ &+\phi_0(\bar{\mathbf{e}})+\frac{1}{2}\Big[\mathbf{F}_{\bar{\mathbf{e}}}\cdot\bar{\mathbb{C}}_H\mathbf{F}_{\bar{\mathbf{e}}}-\fint_Y \mathbf{P}\cdot\mathbb{C}_{\#}^{-1}\mathbf{P}\Big],\end{aligned} \tag{104}$$

which can be regarded as a lower bound. We remark that tighter bounds on the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$ parallel to the classic Hashin-Strikman's bounds can be derived by similar methods used in Liu (2010).

We remark that the rigorous bounds (102) and (104) are implicit and microstructure-dependent since the effective tensors $\mathbb{C}^{\text{eff}}, \boldsymbol{\epsilon}^{\text{eff}}$ and quantities defined in (101) are unknown. Nevertheless, they still provide important benchmarks for numerical computations of effective properties for specified microstructures of electrostrictive composites.

6.2. Effective piezoelectricity of laminated electrets

In this section we address the two-phase simple laminate as shown in Fig. 4 for which closed-form results are possible. The effective electrostrictive tensor for laminates has been explicitly calculated in Tian (2007), Tian et al. (2012) where external charges and dipoles are absent. Earlier works (Deng et al., 2014a, 2014b; Lefevre and Lopez-Pamies, 2017) have addressed the effective piezoelectric responses of electrets without differentiating the apparent and effective piezoelectricity.

Within the rescaled unit cell Y as shown in Fig. 4(b), we assume that the two layers are made of ideal isotropic dielectric elastomers with Lamé constants μ_α, λ_α, electric permittivity ϵ_α, and volume fraction θ_α $(\alpha=1,2)$. Without loss of generality, we assume that the normal to the layers is aligned with $\mathbf{e}_1$-direction and the external polarization on each layer is given by $p_\alpha\mathbf{e}_1$ $(\alpha=1,2)$. From Remark 2, (26), the electrostrictive tensor of the two layers is given by $\mathbb{A}^{(\alpha)}=\frac{\epsilon_\alpha}{2}\mathbb{T}$.

To determine the effective piezoelectricity defined by (69), we first consider the unit cell electrostatic problem (56) that determines the microscopic oscillatory electric field $-\nabla\xi'$:

$$\begin{cases}\frac{d^2\xi'}{dx^2}=0 \qquad \text{if } x\in(0,\theta_1)\cup(\theta_1,1),\\ \Big(-\epsilon_1\frac{d\xi'}{dx}+p_1\Big)\Big|_{\theta_1-}=\Big(-\epsilon_2\frac{d\xi'}{dx}+p_2\Big)\Big|_{\theta_1+},\\ \xi'(0)=\xi'(1).\end{cases}$$

A solution to the above equations implies that

$$-\frac{d\xi'}{dx}=\begin{cases}\theta_2\dfrac{p_2-p_1}{\theta_1\epsilon_2+\theta_2\epsilon_1} & \text{if } x\in(0,\theta_1),\\ -\theta_1\dfrac{p_2-p_1}{\theta_1\epsilon_2+\theta_2\epsilon_1} & \text{if } x\in(\theta_1,1).\end{cases}$$

Next, we need to calculate the electric field $-\nabla\xi_{\bar{\mathbf{e}}}$ associated with an applied average electric field $\bar{\mathbf{e}}\in\mathbb{R}^3$ that is determined by $(31)_1$. For a laminate as shown in Fig. 4(b), $(31)_1$ implies that

$$-\nabla\xi_{\mathbf{e}_1}=\begin{cases}\dfrac{\epsilon_2}{\theta_1\epsilon_2+\theta_2\epsilon_1}\mathbf{e}_1 & \text{if } x\in(0,\theta_1),\\ \dfrac{\epsilon_1}{\theta_1\epsilon_2+\theta_2\epsilon_1}\mathbf{e}_1 & \text{if } x\in(\theta_1,1),\end{cases}$$
$$-\nabla\xi_{\mathbf{e}_2}=\mathbf{e}_2 \text{ in } Y.$$

Thirdly, we consider the elastic unit cell problem $(68)_1$. If $\bar{\mathbf{e}}=\mathbf{e}_1$, the source term in $(68)_1$ is given by

$$2\mathbb{A}_{\#}\left(\nabla\xi'\otimes\nabla\xi_{\mathbf{e}_1}\right)=\begin{cases}\dfrac{\theta_2\epsilon_1\epsilon_2(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)^2}(2\mathbf{e}_1\otimes\mathbf{e}_1-\mathbf{I}) & \text{if } x\in(0,\theta_1),\\ -\dfrac{\theta_1\epsilon_1\epsilon_2(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)^2}(2\mathbf{e}_1\otimes\mathbf{e}_1-\mathbf{I}) & \text{if } x\in(\theta_1,1),\end{cases}\tag{105}$$

which is constant in each of the layers. Therefore, we expect that the strain as determined by $(68)_1$ is piecewise constant and denoted by

$$\nabla\mathbf{u}_1'=\begin{cases}\mathbf{F}_1 & \text{if } x\in(0,\theta_1),\\ \mathbf{F}_2 & \text{if } x\in(\theta_1,1).\end{cases}\tag{106}$$

From the compatibility condition, the equilibrium equation $(68)_1$, and the fact $\fint_Y\nabla\mathbf{u}_1'=0$ we obtain that for some vector $\mathbf{a}\in\mathbb{R}^3$,

$$\begin{aligned}&\mathbf{F}_1-\mathbf{F}_2=\mathbf{a}\otimes\mathbf{e}_1,\\ &(\mathbb{C}_1\mathbf{F}_1-\mathbb{C}_2\mathbf{F}_2)\mathbf{e}_1=-\frac{\epsilon_1\epsilon_2(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)^2}\mathbf{e}_1,\\ &\theta_1\mathbf{F}_1+\theta_2\mathbf{F}_2=0.\end{aligned}\tag{107}$$

From the above equations, we immediately find that

$$\begin{cases}\mathbf{F}_1=\beta_1\theta_2\mathbf{e}_1\otimes\mathbf{e}_1,\\ \mathbf{F}_2=-\beta_1\theta_1\mathbf{e}_1\otimes\mathbf{e}_1,\end{cases}$$
$$\beta_1=-\frac{\epsilon_1\epsilon_2(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)^2[\theta_1(2\mu_2+\lambda_2)+\theta_2(2\mu_1+\lambda_1)]}.$$

Finally, according to the definition $(69)_1$ we conclude that

$$\begin{aligned}\mathbb{B}^{\text{eff}}\mathbf{e}_1&=\fint_Y\left[\mathbb{C}_{\#}(\tilde{\mathbf{x}})\nabla_{\tilde{\mathbf{x}}}\mathbf{u}_1'+2\mathbb{A}_{\#}(\tilde{\mathbf{x}})\left(\nabla_{\tilde{\mathbf{x}}}\xi'\otimes\nabla_{\tilde{\mathbf{x}}}\xi_{\bar{\mathbf{e}}}\right)\right]\\ &=\beta_1\theta_1\theta_2(\mathbb{C}_1-\mathbb{C}_2)\mathbf{e}_1\otimes\mathbf{e}_1\\ &=\beta_1\theta_1\theta_2[2(\mu_1-\mu_2)\mathbf{e}_1\otimes\mathbf{e}_1+(\lambda_1-\lambda_2)\mathbf{I}].\end{aligned}\tag{108}$$

If $\bar{\mathbf{e}}=\mathbf{e}_2$, the source term in $(68)_1$ for is given by

$$2\mathbb{A}_{\#}\left(\nabla\xi'\otimes\nabla\xi_{\mathbf{e}_1}\right)=\begin{cases}\dfrac{\theta_2\epsilon_1(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)}(\mathbf{e}_1\otimes\mathbf{e}_2+\mathbf{e}_2\otimes\mathbf{e}_1) & \text{if } x\in(0,\theta_1),\\ -\dfrac{\theta_1\epsilon_2(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)}(\mathbf{e}_1\otimes\mathbf{e}_2+\mathbf{e}_2\otimes\mathbf{e}_1) & \text{if } x\in(\theta_1,1),\end{cases}$$

Again we consider a solution to $(68)_1$ of form (106). By the compatibility condition, the equilibrium Eq. $(68)_1$, and $\fint_Y\nabla\mathbf{u}_1'=0$, we obtain that for some vector $\mathbf{b}\in\mathbb{R}^3$,

$$\begin{aligned}&\mathbf{F}_1-\mathbf{F}_2=\mathbf{b}\otimes\mathbf{e}_1,\\ &(\mathbb{C}_1\mathbf{F}_1-\mathbb{C}_2\mathbf{F}_2)\mathbf{e}_1=-(p_2-p_1)\mathbf{e}_2,\\ &\theta_1\mathbf{F}_1+\theta_2\mathbf{F}_2=0.\end{aligned}$$

Therefore, the solution to $(68)_1$ is given by

$$\nabla\mathbf{u}_1'=\begin{cases}\beta_2\theta_2\mathbf{e}_2\otimes\mathbf{e}_1 & \text{if } x\in(0,\theta_1),\\ -\beta_2\theta_1\mathbf{e}_2\otimes\mathbf{e}_1 & \text{if } x\in(\theta_1,1),\end{cases}\qquad\beta_2=-\frac{p_2-p_1}{\mu_1+\mu_2}.$$

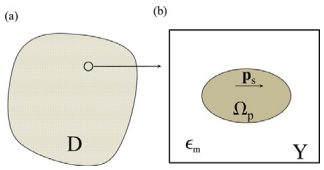

Fig. 5. An electret consisting of a soft matrix with second-phase dipolar inclusions: (a) The overall composite body, (b) a rescaled unit cell.

Finally, by $(69)_1$ we find that

$$\begin{aligned}\mathbb{B}^{\text{eff}}\mathbf{e}_2 &= \beta_2\theta_1\theta_2(\mathbb{C}_1-\mathbb{C}_2)(\mathbf{e}_2\otimes\mathbf{e}_1)+\frac{\theta_2\theta_1(\epsilon_1-\epsilon_2)(p_2-p_1)}{(\theta_1\epsilon_2+\theta_2\epsilon_1)}(\mathbf{e}_1\otimes\mathbf{e}_2+\mathbf{e}_2\otimes\mathbf{e}_1)\\ &= \mathbb{B}^{\text{eff}}_{122}(\mathbf{e}_1\otimes\mathbf{e}_2+\mathbf{e}_2\otimes\mathbf{e}_1).\end{aligned}$$

where

$$\mathbb{B}^{\text{eff}}_{122}=\theta_1\theta_2(p_2-p_1)\Big[\frac{\mu_2-\mu_1}{\mu_2+\mu_1}+\frac{\epsilon_1-\epsilon_2}{(\theta_1\epsilon_2+\theta_2\epsilon_1)}\Big].$$

Remark 7. In practice, it is often more convenient to assess the actuation capability of an electric field by the actuating strain. Associated with the actuation stresses (108) and (109), the strain tensor $\mathbf{S}_{\mathbf{e}_i}\in\mathbb{R}^{3\times3}_{\text{sym}}$ can be defined as (cf., (99))

$$\mathbb{C}^{\text{eff}}\mathbf{S}_{\mathbf{e}_i}+\mathbb{B}^{\text{eff}}\mathbf{e}_i=0, \tag{109}$$

where $\mathbb{C}^{\text{eff}}$ is the effective stiffness tensor of the electret. In particular, if the constituent materials are incompressible, i.e., $\lambda_\alpha\to+\infty$ $(\alpha=1,2)$, the associated actuation strain $\mathbf{S}_{\mathbf{e}_i}$ can be found by solving the following algebraic equations:

$$\mathbb{C}^{\text{eff}}\mathbf{S}_{\mathbf{e}_i}+\eta\mathbf{I}+\mathbb{B}^{\text{eff}}\mathbf{e}_i=0\quad\text{and}\quad \text{Tr}\mathbf{S}_{\mathbf{e}_i}=0, \tag{110}$$

where η is the Lagrange's multiplier associated with the constraint of incompressibility. From (108) and (110), we immediately find that

$$\mathbf{S}_{\mathbf{e}_1}=\left(\mathbb{C}^{\text{eff}}\right)^{-1}\mathbb{B}^{\text{eff}}\mathbf{e}_1$$

whereas

$$\begin{aligned}&\mathbf{S}_{\mathbf{e}_2}=S^{12}(\mathbf{e}_1\otimes\mathbf{e}_2+\mathbf{e}_2\otimes\mathbf{e}_1),\\ &S^{12}=-\frac{\theta_1\theta_2(p_2-p_1)(\theta_1\mu_2+\theta_2\mu_1)}{\mu_1\mu_2}\Big[\frac{\mu_2-\mu_1}{\mu_2+\mu_1}+\frac{\epsilon_1-\epsilon_2}{(\theta_1\epsilon_2+\theta_2\epsilon_1)}\Big].\end{aligned}$$

Remark 8. Our earlier work (Deng et al., 2014b) presented the "effective piezoelectricity" for a finite double layer structure. The result (Eq. (37) in Deng et al. (2014b)) includes the apparent piezoelectricity discussed in Section 5 and the structural effect of double layers, and hence cannot be directly compared with the effective piezoelectricity obtained in Remark 7. Nevertheless, from (105) we may recover Eq. (37) in Deng et al. (2014b) if the associated boundary value problem is solved (within the same approximation).

6.3. Effective piezoelectricity of electrets without elastic contrast or dielectric contrast

In this section, we consider an electret of two-phase materials with single-domain inclusions embedded in a soft matrix. The permittivity and stiffness tensor of the matrix (resp. inclusion) are given by ϵ_m (resp. ϵ_p) and $\mathbb{C}_m$ (resp. $\mathbb{C}_p$). For simplicity, we assume that the inclusions are *uniformly* polarized with polarization $\mathbf{p}_s$, which is a given constant vector and independent of the electric field. As illustrated in Fig. 5, let Y be the rescaled unit cell and $\Omega_p\subset Y$ be the (rescaled) polarized inclusion, and denote by χ_p (resp. χ_m) the characteristic function of the domain Ω_p (resp. $\mathbb{R}^3\setminus\Omega_p$), i.e.,

$$\chi_p=\begin{cases}1 & \text{in } \Omega_p,\\ 0 & \text{in } Y\setminus\Omega_p;\end{cases}\qquad \chi_m=\begin{cases}0 & \text{in } \Omega_p,\\ 1 & \text{in } Y\setminus\Omega_p.\end{cases}$$

To find the effective properties of this electret material, we first need to consider the electrostatic problems, and solve the unit cell problem (56) for $\xi'\in\mathcal{P}_0$ induced by the external polarization $\mathbf{p}_s$:

$$\text{div}\big[-(\epsilon_m\chi_m+\epsilon_p\chi_p)\nabla\xi'+\mathbf{p}_s\chi_p\big]=0, \tag{111}$$

and the unit cell problem $(31)_1$ for $\xi_{\bar{\mathbf{e}}}\in\mathcal{P}_{\bar{\mathbf{e}}}$ associated with the average electric field $\bar{\mathbf{e}}\in\mathbb{R}^3$:

$$\text{div}[-(\epsilon_m\chi_m+\epsilon_p\chi_p)\nabla\xi_{\bar{\mathbf{e}}}]=0. \tag{112}$$

Then we solve the elastic unit cell problem $(68)_1$ for microscopic periodic strain $\nabla\mathbf{u}_1'$:

$$\text{div}\big(\mathbb{C}_\#\nabla\mathbf{u}_1'+\boldsymbol{\sigma}^{(e)}\big)=0, \tag{113}$$

where $\mathbb{C}_{\#} = \mathbb{C}_p\chi_p + \mathbb{C}_m\chi_m$, $2\mathbb{A}_{\#} = \epsilon_{\#}\mathbb{T}$, $\epsilon_{\#} = \epsilon_m\chi_m + \epsilon_p\chi_p$, and

$$\boldsymbol{\sigma}^{(e)} \equiv \epsilon_{\#}\mathbb{T}\big(\nabla\xi' \otimes \nabla\xi_{\bar{\mathbf{e}}}\big). \tag{114}$$

Physically, $\boldsymbol{\sigma}^{(e)}$ can be interpreted as an eigenstress induced by electric fields in the context of Eshelby's inclusions problem (Eshelby, 1957).

According to the definition (69), the effective piezoelectricity tensor is given by

$$\mathbb{B}^{\text{eff}}\bar{\mathbf{e}} = \fint_Y \big[\mathbb{C}_{\#}\nabla\mathbf{u}_1' + \boldsymbol{\sigma}^{(e)}\big]. \tag{115}$$

The boundary value problems (111)–(113) are all linear and can be conveniently solved by a numerical approach. In particular, if there is neither elastic contrast (i.e., $\mathbb{C}_p = \mathbb{C}_m$) nor dielectric contrast (i.e., $\epsilon_p = \epsilon_m$), we immediately see that $\mathbb{B}^{\text{eff}} \equiv 0$ as mentioned in Remark 6. Closed-form expressions for $\mathbb{B}^{\text{eff}}$ seem to be elusive if both elastic and dielectric contrasts exist, except for simple laminates discussed in the last section. However, if one of the contrasts vanishes, we can find closed-form formulae of $\mathbb{B}^{\text{eff}}$ for some special microstructures and some interesting *exact relations* between the effective tensors $\mathbb{B}^{\text{eff}}$, $\mathbb{A}^{\text{eff}}$, $\mathbb{C}^{\text{eff}}$ and $\boldsymbol{\epsilon}^{\text{eff}}$.

6.3.1. No elastic contrast

If there is no elastic contrast, i.e., $\mathbb{C}_p = \mathbb{C}_m$, from (115) we see that

$$\mathbb{B}^{\text{eff}}\bar{\mathbf{e}} = \fint_Y \boldsymbol{\sigma}^{(e)} = \fint_Y 2\mathbb{A}_{\#}\big(\nabla\xi' \otimes \nabla\xi_{\bar{\mathbf{e}}}\big), \tag{116}$$

and hence it is unnecessary to solve the elastic unit cell problem (113). Moreover, comparing (111) and (112), we find that the two solutions are related by

$$-\nabla\xi' = -\nabla\xi_{\bar{\mathbf{e}}} - \bar{\mathbf{e}} \qquad \text{if } \mathbf{p}_s = (\epsilon_p - \epsilon_m)\bar{\mathbf{e}}. \tag{117}$$

By (43), since $\mathbb{C}^{\text{eff}} = \mathbb{C}_m = \mathbb{C}_p$ the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$ satisfies that

$$\mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) = \fint_Y \mathbb{A}_{\#}\nabla\xi_{\bar{\mathbf{e}}} \otimes \nabla\xi_{\bar{\mathbf{e}}}. \tag{118}$$

Setting $\bar{\mathbf{e}} = \mathbf{p}_s/p_s$ ($p_s = |\mathbf{p}_s|$) and comparing (116) and (118), by (117) we see the solution to (116) satisfies

$$\nabla\xi' = \frac{p_s}{\epsilon_p - \epsilon_m}(\nabla\xi_{\bar{\mathbf{e}}} + \bar{\mathbf{e}}).$$

Therefore, by (116) we obtain the following *exact relation* between $\mathbb{B}^{\text{eff}}$, $\mathbb{A}^{\text{eff}}$ and $\boldsymbol{\epsilon}^{\text{eff}}$ when $\bar{\mathbf{e}} = \mathbf{p}_s/p_s$:

$$\begin{aligned}\mathbb{B}^{\text{eff}}\bar{\mathbf{e}} &= \frac{2p_s}{\epsilon_p - \epsilon_m}\fint_Y \mathbb{A}_{\#}[(\nabla\xi_{\bar{\mathbf{e}}} + \bar{\mathbf{e}}) \otimes \nabla\xi_{\bar{\mathbf{e}}}] \\ &= \frac{p_s}{\epsilon_p - \epsilon_m}\big[2\mathbb{A}^{\text{eff}}(\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) - \mathbb{T}\big(\bar{\mathbf{e}} \otimes \boldsymbol{\epsilon}^{\text{eff}}\bar{\mathbf{e}}\big)\big].\end{aligned} \tag{119}$$

Further, taking trace of (116) we obtain

$$\text{Tr}(\mathbb{B}^{\text{eff}}\bar{\mathbf{e}}) = -\fint_Y \nabla\xi' \cdot \epsilon_{\#}\nabla\xi_{\bar{\mathbf{e}}} = 0 \qquad \forall\, \bar{\mathbf{e}} \in \mathbb{R}^3. \tag{120}$$

We emphasize that the exact relations (119) and (120) are valid for any microstructure.

6.3.2. No dielectric contrast

If there is no dielectric contrast, i.e., $\epsilon_p = \epsilon_m$, we see that a solution to the unit cell problem (112) for a constant vector $\bar{\mathbf{e}} \in \mathbb{R}^3$ is given by

$$-\nabla\xi_{\bar{\mathbf{e}}} = \bar{\mathbf{e}} \qquad \text{in } Y,$$

and that a solution to (111) is given by

$$\xi' = \frac{1}{\epsilon_m}\mathbf{p}_s \cdot \nabla u,$$

where u is a Y-periodic potential that satisfies

$$\Delta u = (1 - \theta_1)\chi_p - \theta_1\chi_m \qquad \text{in } Y,$$

and θ_1 is the volume fraction of the inclusion phase. Therefore, the gradient satisfies

$$\begin{aligned}\nabla\xi'|_{\partial\Omega_p^-} &= \frac{1}{\epsilon_m}\mathbf{Q}^-\mathbf{p}_s, \\ \nabla\xi'|_{\partial\Omega_p^+} &= \frac{1}{\epsilon_m}\mathbf{Q}^+\mathbf{p}_s = \frac{1}{\epsilon_m}[\mathbf{Q}^-\mathbf{p}_s - (\mathbf{n} \cdot \mathbf{p}_s)\mathbf{n}],\end{aligned} \tag{121}$$

where $\mathbf{Q} := \nabla\nabla u$ is the second gradient of the potential u, $\mathbf{Q}^- = \mathbf{Q}|_{\partial\Omega_p^-}$ (resp. $\mathbf{Q}^+ = \mathbf{Q}|_{\partial\Omega_p^+}$) represents the interior (resp. exterior) boundary value, $\mathbf{n}$ is the outward unit normal on $\partial\Omega_p$, and the last equality in (121) follows from $[\![\mathbf{Q}]\!]|_{\partial\Omega_p} = -\mathbf{n}\otimes\mathbf{n}$.

Next, we consider the elastic unit-cell problem (113). It is clear that

$$\fint_Y \boldsymbol{\sigma}^{(e)} = \fint_Y 2\mathbb{A}_\#\big(\nabla\xi'\otimes\nabla\xi_{\bar{\mathbf{e}}}\big) = \epsilon_m\mathbb{T}\Big[-\fint_Y \nabla\xi'\otimes\bar{\mathbf{e}}\Big] = 0, \tag{122}$$

and that both ξ' and $\xi_{\bar{\mathbf{e}}}$ are harmonic except at the interface $\partial\Omega_p$, i.e., $\xi'_{,jj}=0$ and $(\xi_{\bar{\mathbf{e}}})_{,jj}=0$ in $\mathbb{R}^3\setminus\partial\bar{\Omega}_p$. Therefore,

$$\sigma^{(e)}_{ij,j} = \epsilon_m\big[\xi'_{,ij}(\xi_{\bar{\mathbf{e}}})_{,j} + \xi'_{,j}(\xi_{\bar{\mathbf{e}}})_{,ij} - \xi'_{,ki}(\xi_{\bar{\mathbf{e}}})_{,k} - \xi'_{,k}(\xi_{\bar{\mathbf{e}}})_{,ik}\big] = 0 \qquad \text{in } Y\setminus\partial\Omega_p. \tag{123}$$

In other words, $\mathrm{div}\boldsymbol{\sigma}^{(e)}=0$ if restricted to the interior or the exterior of the domain Ω_p. Therefore, the eigenstress $\boldsymbol{\sigma}^{(e)}$ is equivalent to an interfacial traction given by

$$\mathbf{t}^{(s)} := [\![\boldsymbol{\sigma}^{(e)}]\!]\mathbf{n}\Big|_{\partial\Omega_p} = \left(\boldsymbol{\sigma}^{(e)}\Big|_{\partial\Omega_p^+} - \boldsymbol{\sigma}^{(e)}\Big|_{\partial\Omega_p^-}\right)\mathbf{n}.$$

We now calculate the interfacial traction $\mathbf{t}^{(s)}$. Restricted to the interior of Ω_p, we have

$$\boldsymbol{\sigma}^{(e)}\Big|_{\partial\Omega_p^-} = -\mathbb{T}\Big[(\mathbf{Q}^-\mathbf{p}_s)\otimes\bar{\mathbf{e}}\Big].$$

Moreover, restricted to the exterior of Ω_p we find that (cf., (121))

$$\boldsymbol{\sigma}^{(e)}\Big|_{\partial\Omega_p^+} = -\mathbb{T}\Big[(\mathbf{Q}^+\mathbf{p}_s)\otimes\bar{\mathbf{e}}\Big] = -\mathbb{T}\Big[(\mathbf{Q}^-\mathbf{p}_s)\otimes\bar{\mathbf{e}} - (\mathbf{n}\cdot\mathbf{p}_s)\mathbf{n}\otimes\bar{\mathbf{e}}\Big].$$

Therefore, the interfacial traction is given by

$$\mathbf{t}^{(s)} = [\![\boldsymbol{\sigma}^{(e)}]\!]\mathbf{n}\Big|_{\partial\Omega_p} = \mathbb{T}\Big[(\mathbf{n}\cdot\mathbf{p}_s)\mathbf{n}\otimes\bar{\mathbf{e}}\Big]\mathbf{n} = (\bar{\mathbf{e}}\otimes\mathbf{p}_s)\mathbf{n} = \boldsymbol{\sigma}^*\mathbf{n}, \tag{124}$$

where $\boldsymbol{\sigma}^* = \bar{\mathbf{e}}\otimes\mathbf{p}_s$. From (123) and (124), we see that the elastic unit-cell problem (113) is equivalent to

$$\mathrm{div}\big(\mathbb{C}_\#\nabla\mathbf{u}'_1 + \boldsymbol{\sigma}^{(e)}\big) = \mathrm{div}\big(\mathbb{C}_\#\nabla\mathbf{u}'_1 - \boldsymbol{\sigma}^*\chi_p\big) = 0. \tag{125}$$

Upon solving the above equation with periodic boundary conditions, we can define a linear mapping $\mathbb{D}:\mathbb{R}^{3\times3}\to\mathbb{R}^{3\times3}$ such that

$$\mathbb{D}\boldsymbol{\sigma}^* = \frac{1}{1-\theta_1}\fint_{\Omega_p}\nabla\mathbf{u}'_1. \tag{126}$$

Then from the definition (69) we find that

$$\begin{aligned}\mathbb{B}^{\mathrm{eff}}\bar{\mathbf{e}} &= \fint_Y\Big[\mathbb{C}_\#\nabla\mathbf{u}'_1 + \boldsymbol{\sigma}^{(e)}\Big] = \fint_Y\Big\{\mathbb{C}_m\nabla\mathbf{u}'_1 + \Big[(\mathbb{C}_p-\mathbb{C}_m)\nabla\mathbf{u}'_1\Big]\chi_p\Big\}\\ &= (\mathbb{C}_p-\mathbb{C}_m)\fint_Y(\nabla\mathbf{u}'_1)\chi_p = \theta_1(1-\theta_1)(\mathbb{C}_p-\mathbb{C}_m)\mathbb{D}\boldsymbol{\sigma}^*,\end{aligned} \tag{127}$$

where $\mathbb{C}_m$ (resp. $\mathbb{C}_p$) is the stiffness tensor of the matrix (resp. inclusion), the second equality follows from (122), and the last equality follows from (126).

Finding the tensor $\mathbb{D}$ defined in (126) in general requires solving the periodic *inhomogeneous* Eshelby inclusion problem (125). There are two scenarios that are of particular interest.

1. If $\boldsymbol{\sigma}^*\in\mathbb{R}^{3\times3}_{\mathrm{sym}}$ and there is an average symmetric strain $\bar{\mathbf{H}}\in\mathbb{R}^{3\times3}_{\mathrm{sym}}$ such that $(\mathbb{C}_p-\mathbb{C}_m)\bar{\mathbf{H}} = -\boldsymbol{\sigma}^*$, then (125) is equivalent to

$$\mathrm{div}\Big[\mathbb{C}_\#\big(\nabla\mathbf{u}'_1+\bar{\mathbf{H}}\big)\Big] = \mathrm{div}\Big[\mathbb{C}_\#\nabla\mathbf{u}'_1 + \chi_p(\mathbb{C}_p-\mathbb{C}_m)\bar{\mathbf{H}}\Big] = 0.$$

From the definition of the effective stiffness tensor $(30)_2$, we have

$$\begin{aligned}\mathbb{C}^{\mathrm{eff}}\bar{\mathbf{H}} &= \fint_Y\mathbb{C}_\#(\nabla\mathbf{u}'_1+\bar{\mathbf{H}}) = \fint_Y\chi_p(\mathbb{C}_p-\mathbb{C}_m)\nabla\mathbf{u}'_1 + \bar{\mathbb{C}}\bar{\mathbf{H}}\\ &= \theta_1(1-\theta_1)(\mathbb{C}_p-\mathbb{C}_m)\mathbb{D}\boldsymbol{\sigma}^* + \bar{\mathbb{C}}\bar{\mathbf{H}}.\end{aligned}$$

Therefore, by (127) we find that

$$\begin{aligned}\mathbb{B}^{\mathrm{eff}}\bar{\mathbf{e}} &= \theta_1(1-\theta_1)(\mathbb{C}_p-\mathbb{C}_m)\mathbb{D}\boldsymbol{\sigma}^* = (\mathbb{C}^{\mathrm{eff}}-\bar{\mathbb{C}})\bar{\mathbf{H}}\\ &= -(\mathbb{C}^{\mathrm{eff}}-\bar{\mathbb{C}})(\mathbb{C}_p-\mathbb{C}_m)^{-1}\boldsymbol{\sigma}^*.\end{aligned}$$

In other words, if $\bar{\mathbf{e}} = \mathbf{p}_s/p_s$, the effective piezoelectric response is directly related with the effective elasticity tensor as

$$\mathbb{B}^{\mathrm{eff}}\bar{\mathbf{e}} = -p_s(\mathbb{C}^{\mathrm{eff}}-\bar{\mathbb{C}})(\mathbb{C}_p-\mathbb{C}_m)^{-1}(\bar{\mathbf{e}}\otimes\bar{\mathbf{e}}). \tag{128}$$

The above closed-form formula relating the effective piezoelectricity tensor and effective elasticity tensor can be regarded as an *exact relation* which is quite extraordinary (cf.Milton, 2002). We refer the reader to the following references and citations therein for a discussion on exact relations in the context of conventional piezoelectricity: (Benveniste, 1994; Benveniste and Milton, 2004).

In the case that the microstructure is a simple laminate along $\bar{\mathbf{e}}$-direction, we can explicitly find $\mathbb{C}^{\text{eff}}$, and hence the effective piezoelectricity tensor as (μ_1, λ_1, θ_1 (resp. μ_2, λ_2, θ_2) are the Lamé constants and volume fraction of the particle (resp. matrix phase))

$$\mathbb{B}^{\text{eff}}\bar{\mathbf{e}} = \frac{-p_s\theta_1\theta_2[2(\mu_1-\mu_2)\mathbf{e}_1\otimes\mathbf{e}_1+(\lambda_1-\lambda_2)\mathbf{I}]}{\theta_1(2\mu_2+\lambda_2)+\theta_2(2\mu_1+\lambda_1)},$$

which agrees with our earlier result (108).

2. In the dilute limit, we assume the domain Ω_p is of the shape of an ellipsoid and the unit cell problem is well approximated by the model of a single inclusion embedded in an infinite matrix. As is well-known in elasticity, the inhomogeneous problem (125) admits a closed form solution, namely, the Eshelby-Walpole's solution. For the coordinate system aligned with the principle axes of the ellipsoid, the $\mathbb{D}$-tensor is explicitly given by Eshelby (1957), Walpole (1991), Liu et al. (2006):

$$\mathbb{D} = \left(\mathbb{S}^{-1}+\mathbb{C}_p-\mathbb{C}_m\right)^{-1}, \tag{129}$$

$$\mathbb{S}_{piqj} = \frac{1}{4\pi}\int_{S^2}\frac{\det(\Lambda)N_{pq}\left(\hat{\mathbf{k}}\right)\hat{k}_i\hat{k}_j}{|\Lambda\hat{\mathbf{k}}|^3}d\hat{\mathbf{k}},$$

where S^2 is the unit sphere in $\mathbb{R}^3$, $N_{pq}(\hat{\mathbf{k}})$ is the inverse matrix of the matrix $(\mathbb{C}_m)_{piqj}\hat{k}_i\hat{k}_j$, i.e., $N_{pr}(\hat{\mathbf{k}})L_{riqj}\hat{k}_i\hat{k}_j=\delta_{pq}$, and $\Lambda = diag[a_1,a_2,a_3]$ (a_1, a_2, a_3 is the half axis-lengths of the ellipsoid Ω_p). From (129) we can immediately obtain the effective piezoelectricity tensor by (127).

7. Summary of the key observations and insights

We have presented a careful study of the effective electroelastic properties of electret materials–materials that have embedded immobile external charges and/or dipoles. We briefly summarize our results below:

1. Electret materials exhibit a measurable piezoelectric response as has been demonstrated by several experimental works. We have highlighted that only part of that response reflects a true "effective piezoelectric" property and should be distinguished from a separate contribution, termed "apparent" piezoelectricity that depends on the structural boundary conditions and is not a true thermodynamically defined bulk property. We provide general relations for both the effective and apparent piezoelectric response of electrets which can now serve as the basis for future numerical calculations and approximate analytical solutions for specific microstructures.
2. We establish rigorous variational bounds for electret materials that can serve as important benchmark for future numerical computations.
3. For the case of laminate microstructure, we obtain explicit closed-form expressions for the effective electromechanical properties of electrets.
4. We find that, for an "effective" piezoelectric response, heterogeneity in either the elastic or dielectric properties is necessary. This is not the case for the contribution of the "apparent" piezoelectricity.
5. Very few results exist in the theory of composites that are considered *exact* i.e. microstructure-independent. We find some remarkable (and unexpected) exact results in the theory of effective properties of electrets if either the elastic or dielectric contrast vanish.
6. Although the coupled problem we have considered is nonlinear, we have worked within the small-deformation theory. Consideration of large deformation is a rather challenging endeavor—at least analytically—and should be an interesting future research avenue.

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