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Magneto-Electro-Mechanical Coupling Mechanisms in Soft Materials

A Dissertation

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in Materials Sciences and Engineering

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Magento-Electro-Mechanical Coupling Mechanisms in Soft Materials

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Abstract

Soft materials are everywhere around and inside us. Because of their abundance, low cost and ease of fabrication along with interesting physical properties, they have numerous promising applications such as artificial muscles, sensors and actuators, smart materials and energy harvesters to name a few. Soft materials deform easily under the presence of external stimuli due to their low mechanical stiffness. This response makes them ideal candidates for the design of multifunctional smart materials. Recently, it was shown that soft materials can produce apparent piezoelectric and magnetoelectric behavior even in the absence of those intrinsic properties provided certain conditions apply. In this dissertation, we will highlight some characteristics and implications of magneto-electro-elastic coupling behavior in specific soft materials structures:

1. The emergence of apparent magnetoelectric behavior in soft materials and its stability:
We explore the interplay between elastic strain, electric voltage and magnetic field and its effect on the maximum stretch and voltage that the material can sustain. We present physical insights to support the design of wireless energy harvesters that can be remotely activated with an external magnetic field.
2. Engineering concurrent magnetoelectricity and piezoelectricity in soft materials using electret structure: We prove that by embedding charges in an elastically heterogeneous soft dielectric structure, it is possible to obtain simultaneous piezoelectricity and magnetoelectricity even in the absence of these intrinsic properties. We show that the coupling coefficients in this case are large and compare to some of the well-known ceramic composites.
3. Enhanced electromagnetomechanical response in solid and liquid inclusions: We design a composite made of a dielectric elastomer as the matrix and a spherical inclusion made of iron or ferrofluid while accounting for capillary effects. The beauty of this composite resides on its ability to respond to external electric and magnetic stimuli. We investigate the effect of surface energy at the inclusion/matrix interface on the effective response of this composite.

4. Microscopic mechanisms underpinning flexoelectricity in soft materials: We prove that the existence of frozen dipoles and their thermal fluctuations contribute to the flexoelectric response of the dielectric material. We also predict a temperature dependence of the coupling coefficient.

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Chapter 1

Introduction

1.1 Magnetoelectricity in soft materials

Magnetoelectric effect was predicted as early as 1894. The ability to control magnetization by an external electric field or polarization by a magnetic field has promising technical applications in the fields of microwave devices, sensors and memory bits. Magnetoelectric materials were discovered in 1950's, and since that date, the quest for material exhibiting a strong ME effect started. Intensive research work was put during the past decades in order to study magnetoelectric coupling in single-phase materials. However, it came disappointing that even though the ME effect is intrinsic in some rare materials, the realizable coupling constant was of the order $1 \sim 20$ mV/cm.Oe; too small to be used effectively in device fabrication. Moreover, most of these materials have Curie or Neel temperature far below the room temperature. Growth of good crystals also represented a major challenge since it calls for expensive materials and processing techniques [1]. As a result, the interest in ME effect declined, and all the applications which were foreseen at the time of its discovery were regarded as unpromising [2].

The revival of this research field came in the 1990's after an alternative approach for engineering magnetoelectricity. The idea consisted of synthesizing a composite medium where the ME effect can be realized by coupling magnetic and electric degrees of freedom indirectly through a third parameter such as mechanical deformation. Combining piezomagnetic and

piezoelectric phases or magnetostrictive and piezoelectric phases can yield the desired property. Typically, in these composites, an external magnetic field creates mechanical strain, which will be transferred to the adjacent piezoelectric material. This, in turn, will generate an electric field because of the stress applied. This way, using product properties, the ME effect can be generated from materials which initially do not allow ME effect [3].

The earliest experiments on granular ME composites revealed a ME coupling constant up to 130 mV/cm.Oe which exceeded the values observed for single-phase materials [4]. Later, the possibility of fabricating laminate composites suitable for practical magnetic noise sensing applications was investigated [1]. Using PZT and Terfenol-D as piezoelectric and magnetostrictive materials respectively, a magnetoelectric voltage coefficient that is about 36 times higher than the best reported values was measured [5]. Despite the major advances in the fabrication of laminate ME composites; many limitations still pose several challenges. In these composites, the magnetoelectric coupling constant would depend on the individual piezoelectric and magnetostrictive coefficients of each material, which in turn are affected by numerous factors. The fabrication process, defects and thermal fluctuations in addition to the mechanical properties of these materials, which are usually brittle ceramics with their disadvantage of small deformation, are factors that affect the engineering of these composites.

Indeed, a universal mechanism was reported recently which applies to all soft polymers and saves the complexity and cost of creating composites ME materials [6]. Soft dielectric materials exhibit a giant magnetoelectric effect although it is neither intrinsically piezoelectric nor magnetostrictive. Because of this strain mediated mechanism, soft polymer foams are predicted to have ME coefficient comparable to the best known hard ME composite created.

1.2 Engineering Apparent Piezoelectricity in Soft Electret Structures

Piezoelectricity is the development of electric polarization under the application of a mechanical strain. This is a two way effect, in the sense that an external electric volt-

age would cause the material to deform. Piezoelectricity requires the material to have non-centrosymmetry [7]. This constraints the presence of this property to a limited set of crystalline materials and very few soft dielectrics. Materials that show intrinsic large piezoelectricity, such as barium titanate and zinc oxide, are very hard ceramics that are not suitable for soft materials application. On the other hand, piezoelectric polymers have considerably smaller response when compared to crystalline materials. PVDF, for example, is one of the very few soft materials that show piezoelectricity [8].

Mathematically, piezoelectricity is defined through the following relation between electric polarization and strain:

$$P_i \sim d_{ijk}\epsilon_{jk}, \quad (1.2.1)$$

where P_i is the induced polarization, ϵ_{jk} is the component of the strain tensor. The piezoelectric coefficients are given by the third order tensor d_{ijk} . Since all odd order tensors vanish under inversion symmetry, we confirm that centrosymmetric materials such as NaCl are not piezoelectric. This form of electromechanical coupling has found wide range of applications in biology, energy harvesting, sensing and actuation in addition to others.

There exists also another electromechanical coupling that is common in all soft materials. Subject to an external electric voltage, all dielectric materials deform. Some expand in area and become thinner, whereas others shrink and become thicker. This mechanism is known as the Maxwell's stress effect. The magnitude of deformation depends significantly on the elastic modulus of the material itself and the field applied. Maxwell's stress is a one way effect. In other words, stretching or squeezing the dielectric material does not result in induced polarization. Moreover, changing the direction of the electric does not change the deformation. Mathematically, this can be understood from the quadratic dependence of the Maxwell's stress on the magnitude of the electric field $\sim \epsilon_0|\mathbf{E}|^2$.

The Maxwell's stress effect is particularly interesting. It allows the possibility to engineer soft dielectric materials that exhibit apparent piezoelectric behavior without being intrinsically so. This can be realized using the concept of electrets. The latter are soft materials composites made by inserting layers of charges or dipoles in an elastically heterogeneous

structure. For instance, multilayers of different dielectric materials with charges embedded at the interface or foamy polymers with charges at the void location satisfy this condition. Experiments have revealed that using some polymer materials, electret structures can be achieved and they are reasonably stable. Under the application of an external voltage, the electret deforms because of the Maxwell's stress. This results in a change in the preexisting polarization in the material. The converse effect also exists. As a result, the electret composite behaves as a piezoelectric material for practical purpose. In fact, piezoelectric coupling coefficient in electret structures such as polypropylene foams are larger than those reported for ceramic composites (such as PZT). Engineering apparent piezoelectricity in soft materials is very promising since these structures offer flexibility and strong coupling behavior which makes them excellent candidate for device fabrication.

1.3 Flexoelectricity

Flexoelectricity denotes the creation of electric polarization due to macroscopic strain gradient. It is a universal mechanism that is common to all materials even those who are centrosymmetric and do not exhibit piezoelectricity. The fundamental difference between flexoelectricity and piezoelectricity can be explained in Fig.1.3.1. If we apply a uniform me-

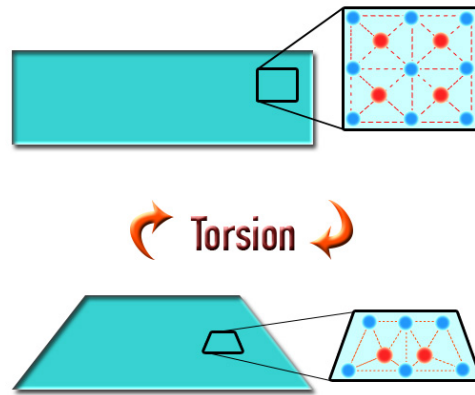


Figure 1.3.1: Effect of a strain gradient on a centrosymmetric material

chanical strain on a centrosymmetric material, there will be no induced polarization because of the symmetry since the centers of positive and negative charges coincide. However, if we apply a non-uniform strain, the centers of the positive and negative charges will shift. As a result, dipoles are created and an induced electric polarization would be detected. In order to account for change of electric polarization due to strain gradient, we extend eq.(1.2.1) to

$$P_i \sim d_{ijk}\epsilon_{jk} + f_{ijkl}\frac{\partial\epsilon_{jk}}{\partial x_l}. \quad (1.3.1)$$

f_{ijkl} is the fourth order flexoelectric tensor.

The flexoelectric effect is well illustrated by non-uniform straining of graphene nanoribbons [9]. In addition, experiments have measured flexoelectricity in several crystalline materials such as barium titanate, polymers, 2D materials as well as biological membranes. Since strain gradient depends on the size of the material, this mechanism is particularly significant because it allows size dependent electromechanical properties at the nanoscale. Researchers have advocated the ability to create artificial piezoelectric materials even in the absence of intrinsic piezoelectricity [10, 11]. Flexoelectric effect can also be exploited to design energy harvesters, sensors and actuators, along with other applications.

1.4 Stability of dielectric elastomers under the application of external stimuli

The presence of large deformations in soft materials constitutes a perfect environment for growing instabilities. It was previously believed that instabilities are unfavorable and destructive. As a result, they were often avoided. More recently however, it was shown that instabilities in soft materials can be exploited and harnessed to enhance materials' response [12, 13, 14]. Researchers have focused on instability modes in a dielectric subject to mechanical loading and electric voltage or magnetic field. It is crucial to understand the conditions of instabilities to suppress them, whenever possible, and avoid device failure. For example, it was shown that dielectric elastomer can withstand electric voltage up

to a certain threshold before the pull in instability occurs. Still it is possible to suppress this instability simply by pre-stress. As a result, the dielectric material can sustain higher voltage, which is critical when it comes to designing actuators and sensors [15, 14]. Also, more energy could be harvested in this case by harnessing large deformations that occur at this critical voltage [12].

The various instability modes of a dielectric elastomer under the application of an electric voltage and mechanical strain has been investigated by several researchers [16, 17, 15, 18, 19, 20, 21]. Some of the modes studied in those references include but not limited to thinning and pull-in instabilities, electro-creasing to cratering, electro-cavitation and wrinkling. On the other hand, the stability of magnetically responsive materials was addressed by few researchers only [22, 23, 24, 25].

1.5 Solid and Liquid Inclusion

Composites are at the heart of natural and engineered materials. It has been experimentally shown that performance of composite materials significantly improves over their individual constituents [26, 27, 28]. Eshelbey has laid theoretical foundations to explore properties of solid composites [29]. His formulation describes the effect of microscopic inclusions on the overall macroscopic stress field and the mechanical properties of the composite itself. Originally, this theory was designed to find the stiffness of dilute solid composites [29, 26]. It has been later extended to study fractures [30] and plasticity [31].

However, one limitation of Eshelbey’s inclusion theory is that it does not properly account for the interface between the matrix and the inclusion. Experiments done on micro and nanosized inclusions have revealed some counterintuitive results [32, 26]. Inserting very soft and small inclusions in a soft materials make the overall composite stiffer. The root cause of this unexpected behavior resides in the effect of surface stress on the effective properties of the material. This is particularly crucial for small inclusions.

The desire to create soft material structure that can reveal strong electromechanical and magnetoelectric coupling behavior has been the main drive behind the growing attention

received by solid and liquid inclusions in soft elastomers. The goal is clear: designing a soft dielectric structure that respond to electric voltage and magnetic field with large deformation. Various exciting technological applications such as robotics, stretchable electronics and energy harvesters to name a few, are on the horizon. Even though dielectric elastomers are capable of large deformation, they have low electric permittivity and magnetic permeability. As a result, they require a very high electric field to be actuated, and they barely respond to a magnetic field. Why are inclusions so important? Well, we expect that inserting high permittivity and permeability finite inclusion in a soft dielectric elastomer would definitely modify the mechanical, electric and magnetic properties of the overall composite. Thus, the composite material will respond to electric voltage and magnetic field while showing large deformation.

1.6 Motivation

Recently, coupling mechanisms which allow conversion between different forms of energy have attracted immense attention due to their role in several promising applications such as artificial muscles, sensors and actuators, energy harvesters and memory devices to name few. Specifically, piezoelectricity, flexoelectricity and magnetoelectricity are some phenomena that have been thoroughly investigated. The quest for a material that has high coupling coefficient suitable for these technological applications is never ending. As a matter of fact, this road has been rocky since intrinsic properties exist only in very rare, often hard, materials. Meanwhile, interest in soft materials is rising because of their attractive mechanical properties such as ease of fabrication, lightweight and flexibility. Moreover, soft materials are readily available and can mimic biological tissues. As such, multifunctional soft materials can pave the way for several tantalizing applications such as soft robots, stretchable electronics and smart skins.

1.7 Outline of The Dissertation

In this thesis, we focus on several aspects related to coupling mechanisms in soft materials. We first study the stability of a soft dielectric material undergoing electric, magnetic

and mechanical stimuli. We explore the interplay between those three parameters and suggest a simple idea of energy harvesting device based on this idea.

Next, we present a simple mechanism to design a composite soft material that exhibit concurrent magnetoelectricity and piezoelectricity using the concept of electrets. In the fourth chapter, we discuss another composite structure which consists of a matrix and an inclusion (liquid or solid) undergoing an electric voltage and an external magnetic field.

In chapter five, we show another interesting microscopic mechanism that explains flexoelectricity in soft materials. We examine how the presence of frozen dipoles impacts the flexoelectric behavior of soft dielectrics. We also come up with a prediction on the dependence of flexoelectricity on temperature. Finally, in the last chapter, we summarize our work and describe future directions that open up due to our research.

Chapter 2

Emergent magnetoelectricity in soft materials, instability, and wireless energy harvesting

Magnetoelectric materials that convert magnetic fields into electricity and vice-versa are rare and usually complex, hard crystalline alloys. Recent work has shown that soft, highly deformable magnetoelectric materials may be created by using a strain-mediated mechanism. The electromagnetic and elastic deformation of such materials is intricately coupled; giving rise to a rather rich instability and bifurcation behavior that may limit or otherwise put bounds on the emergent magnetoelectric behavior. In this chapter, we investigate the magneto-electro-mechanical instability of a soft dielectric film subject to mechanical forces and external electric and magnetic fields. We explore the interplay between mechanical strain, electric voltage and magnetic fields and their impact on the maximum voltage and the stretch the dielectric material can reach. Specifically, we present physical insights to support the prospects to achieve wireless energy harvesting through remotely applied magnetic fields.

2.1 Introduction

Soft materials that are capable of large mechanical deformation in response to an external stimuli (such as temperature, pH or electric field) have recently received extensive attention. As example, soft *dielectrics* respond to electric fields and have been investigated for applications in human-like robots [33, 34], stretchable electronics [35], actuators [36, 14, 37] and energy harvesting [13, 12, 38, 39, 40]. Magnetoelectric coupling refers to the ability of the material to electrically polarize under the application of a magnetic field, and conversely, magnetize under the application of an electric field. Unfortunately, there are no known *natural* soft magnetoelectric materials. Magnetoelectricity was discovered in a class of hard, crystalline multiferroics [41, 42, 43]. The intrinsic coupling is low at room temperature and these materials can hardly sustain large deformations¹.

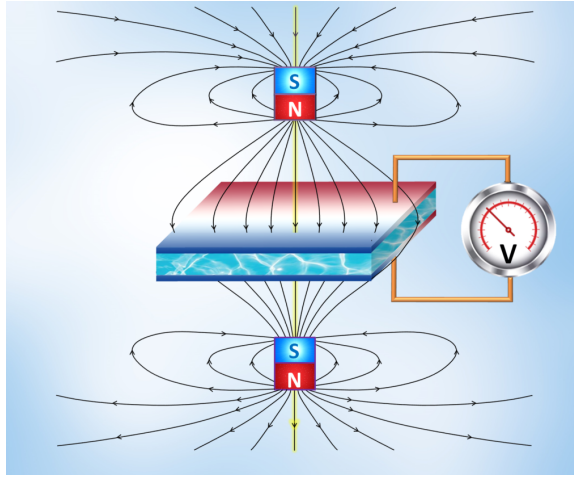


Figure 2.1.1: Schematic figure illustrating the mechanism that can be used to engineer the magnetoelectric effect in soft materials.

Soft materials that are magnetoelectric are expected to have several interesting applications such as wireless energy transfer [46], spintronics and nonvolatile memories [47], multiple state energy bits that can be written electrically and retrieved magnetically, among others [43]. Perhaps the most enticing one is that of wireless energy harvesting. Magnetic

¹An alternative approach to artificially design hard composite magnetoelectric materials is by combining piezoelectric and magnetostrictive materials. Most such resulting materials are also hard, the ensuing coupling not strong and suffer from challenges related to cost-effectiveness and the associated complex fabrication processes. [44, 45]

fields may be remotely imposed and therefore a suitable magnetoelectric soft material may provide a facile route to convert magnetic power into electric energy. Coupled with the large-deformation capability of soft materials, these materials present a compelling case as actuators, sensors and energy conversion devices. In recent works, a rather simple approach to create artificial soft magnetoelectric materials was proposed by Liu, Sharma and co-workers [6, 48, 49] that does not require the materials themselves to be magnetoelectric, or piezoelectric or exhibit any exotic atomistic features that conventional hard crystalline multiferroics do. Rather, any soft material may be made to act like a magnetoelectric material (Fig.2.1.1) provided certain conditions are met.

For instance, a soft dielectric film coated with two compliant electrodes under an applied voltage will deform because of the electric Maxwell stress, and this deformation is proportional to the square of the applied electric field. What will happen to the dielectric response if an external magnetic field is applied in addition to the applied voltage? *If* the dielectric material *also* has a magnetic permeability larger than unity, then it will deform *further* due to the magnetic Maxwell stress. The superimposed additional deformation will, in turn, alter the pre-existing electric field and thus, change the polarization (see Fig.2.1.1). The detectable change in the electric field resulting from the application of the magnetic field manifests itself as a magnetoelectric effect [6, 48]. Since the aforementioned mechanism relies mainly on electric and magnetic Maxwell stress the resulting magnetoelectric effect is universal. Here, we note three essential conditions for the effect to take place [6, 48, 49]. First, the material must be mechanically soft so that the electrical Maxwell stress effect is significant. Second, the dielectric material must have a magnetic permeability larger than that of vacuum. That is, the magnetic permeability of the soft material must be greater than one, $\mu_r > 1$. The latter can be ensured by incorporating a very minute amount of soft magnetic particles or fluid [24]. Finally, a pre-existing electric field must be present. Large deformation in soft matter and instabilities go hand-in-hand. For example, soft dielectrics are vulnerable to a wide range of electro-mechanical instabilities including thinning and pull-in instabilities [16, 17, 15], electro-creasing to cratering [18], electro-cavitation [19] and wrinkling to name just a few [20, 21]. Historically, instabilities have been consid-

ered harmful (and they indeed can be) but more recently, especially in the context of soft dielectrics, they have also been exploited to enhance material behavior and design novel devices [50]. While the literature on the discussion of instability in dielectrics (and its avoidance or enhancement) is extensive, very few works have focussed on analogous issues in the context of soft magnetically responsive materials [22, 23, 24, 25]. In this chapter, (i) we analyze the magneto-electro-mechanical stability of a soft dielectric film subjected to a combination of mechanical, electrical and magnetic stimuli, (ii) present insights into the resulting phenomenon of strain-mediated magnetoelectricity, and (iii) explore the prospects for wireless energy harvesting due to remotely applied magnetic fields.

The outline of this chapter is as follows: In Section 2.2, we present the theoretical formulation, first in general terms and then specialized for the thin-film configuration of interest which we use to illustrate our key results. In Section 2.3 we specialize our derivations to homogeneous deformation and for an ideal dielectric elastomer. We present the central results related to stability and magnetoelectric effect in Section 2.4 and discuss wireless energy harvesting in Section 2.5. Finally, we conclude our work in Section 2.6.

2.2 Formulation

In this section, we derive the governing equations needed to investigate the magneto-electromechanical stability of soft dielectric materials subjected to a combination of mechanical forces and external electric and magnetic fields. We assume the simplest possible configuration: a film of soft material sandwiched between two electrodes (see Fig.2.2.1). This will facilitate exploration of the key mechanisms and insights underpinning magnetoelectricity in soft materials while avoiding excessive mathematical tedium. The material forming the layer is assumed elastically nonlinear but electrostatically and magnetostatically linear.

2.2.1 Geometry and deformation

If we consider a film of soft dielectric and choose a positively-oriented, orthonormal basis $\{\mathbf{e}_X, \mathbf{e}_Y, \mathbf{e}_Z\}$ with associated Cartesian coordinates X , Y , and Z , the domain occupied by the dielectric film (see Fig.2.2.1), in the reference configuration, is given by

$$\mathcal{B} = \{\mathbf{X} \in \mathbb{R}^3 : 0 \leq X \leq L_1, 0 \leq Y \leq L_2, 0 \leq Z \leq L_3\}, \quad (2.2.1)$$

where $L_i, i = 1, 2, 3$, are geometrical dimensions.

The deformation is denoted by a smooth mapping: $\chi : \mathcal{B} \rightarrow \mathbb{R}^3$ and the deformed film dimensions become (l_1, l_2, l_3) . In contrast to the material point $\mathbf{X} \in \mathcal{B}$, the spatial point is then represented by $\mathbf{x} = \chi(\mathbf{X})$, which is denoted by the Cartesian triplet (x, y, z) in the current configuration.

We consider the following class of deformations

$$x = X + a(X), \quad y = Y(1 + b(X)), \quad z = Z(1 + c(X)), \quad (2.2.2)$$

where $a(X)$, $b(X)$ and $c(X)$ are functions of only the variable X .

The deformation gradient is

$$\mathbf{F} = \frac{\partial \chi}{\partial \mathbf{X}} = \begin{pmatrix} \lambda_1 & 0 & 0 \\ Yb'(X) & \lambda_2 & 0 \\ Zc'(X) & 0 & \lambda_3 \end{pmatrix}, \quad (2.2.3)$$

where the prime denotes the derivative with respect to X and the stretches² are

$$\lambda_1 = \frac{\partial x}{\partial X} = 1 + a'(X), \quad (2.2.4a)$$

²The stretches here are not the principal stretches of the deformation.

$$\lambda_2 = \frac{\partial y}{\partial Y} = 1 + b(X), \quad (2.2.4b)$$

and

$$\lambda_3 = \frac{\partial z}{\partial Z} = 1 + c(X). \quad (2.2.4c)$$

The (volumetric) Jacobian then becomes

$$J = \det \mathbf{F} = \lambda_1 \lambda_2 \lambda_3, \quad (2.2.5)$$

where λ_i is given in (2.2.4). For incompressible materials, the Jacobian in (2.2.5) is unity

$$J = 1 \quad (2.2.6)$$

and then the stretches have the relation

$$\lambda_1 = \frac{1}{\lambda_2 \lambda_3}. \quad (2.2.7)$$

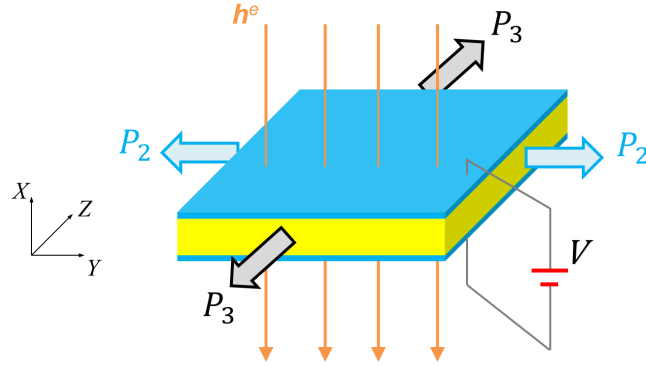


Figure 2.2.1: A film of soft dielectric material subject to two pairs of in-plane mechanical loads P_2 and P_3 , an applied electrical voltage difference V , and an external magnetic field h^e in the thickness direction.

2.2.2 Maxwell's equations and boundary conditions

Maxwell's equations

In the current configuration, the Maxwell equations are [51, 48]

$$\operatorname{div}(-\epsilon_0 \operatorname{grad} \xi + \mathbf{p}) = 0, \quad \operatorname{div}(-\operatorname{grad} \zeta + \mathbf{m}) = 0, \quad (2.2.8)$$

where ϵ_0 is the vacuum permittivity, “div” and “grad” are the divergence and gradient operators, ξ is the electric potential field and ζ is the magnetic potential field. $\mathbf{p}$ and $\mathbf{m}$ respectively denote the polarization and the magnetization.

The relation between the polarization and the magnetization in the current and reference configurations are [6, 51, 48]

$$\mathbf{p} = \frac{\mathbf{P}}{J}, \quad \mathbf{m} = \frac{\mathbf{M}}{J}, \quad (2.2.9)$$

where $\mathbf{P}$ and $\mathbf{M}$ respectively denote the polarization and the magnetization in the reference configuration. Other definitions of these relations between the true and the nominal polarization (or magnetization), see for example the definition by Dorfmann and Ogden [52], can also be used if the expressions of the corresponding Maxwell equations are properly defined. The current choice of the defined nominal magnetization in (2.2.9) just makes consistent definitions of both the nominal polarization and the nominal magnetization. Properly defined expressions of the Maxwell equations are necessary.

Electric and magnetic boundary conditions

The electric boundary conditions on the upper ($x = l_1$) and bottom ($x = 0$) surfaces in the current configuration are

$$\xi|_{x=l_1} = V, \quad \xi|_{x=0} = 0, \quad (2.2.10)$$

where x is the coordinates in the current configuration and l_1 is the thickness of the deformed film.

The far-field magnetic boundary condition is [51, 48]

$$-\text{grad } \zeta \rightarrow \mathbf{h}^e \quad \text{as } |\mathbf{x}| \rightarrow \infty, \quad (2.2.11)$$

where $\mathbf{x}$ is the spatial point and $\mathbf{h}^e = h^e \mathbf{e}_x$ is the external far-field magnetic field in the current configuration with basis $\{\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z\}$.

From Ampère's law for time-independent problems and in the absence of external currents, we have the following interface discontinuity/boundary condition for the magnetic field on the upper ($x = l_1$) and bottom ($x = 0$) surfaces

$$\mathbf{n} \times \llbracket -\text{grad } \zeta \rrbracket = \mathbf{0} \quad \text{on } x = 0 \text{ \& } l_1, \quad (2.2.12)$$

where $\mathbf{n}$ is a unit normal to the surface and $\llbracket f \rrbracket = f_+ - f_-$, is the difference of the field quantity f evaluated at either side of the discontinuity surface.

In addition, given the absence of “magnetic monopoles” on the interfaces, we have the following interface discontinuity/boundary condition for the magnetic flux:

$$\mathbf{n} \cdot \llbracket -\mu \text{grad } \zeta \rrbracket = 0 \quad \text{on } x = 0 \text{ \& } l_1, \quad (2.2.13)$$

where μ is the magnetic permeability.

2.2.3 Free energy of the system

The total free energy of a general conservative magneto-electro-mechanical system in a three-dimensional space can be expressed as [51]

$$\mathcal{F}[\chi, \mathbf{P}, \mathbf{M}] = \mathcal{U}[\chi, \mathbf{P}, \mathbf{M}] + \mathcal{E}^{\text{elect}}[\chi, \mathbf{P}] + \mathcal{E}^{\text{mag}}[\chi, \mathbf{M}] + \mathcal{P}^{\text{mech}}[\chi]. \quad (2.2.14)$$

Here, $\mathcal{U}$ is the internal energy

$$\mathcal{U} = \int_{\mathcal{B}} W(\chi, \mathbf{P}, \mathbf{M}), \quad (2.2.15)$$

where $\mathcal{B}$ is the domain of the system in the reference configuration and $W(\chi, \mathbf{P}, \mathbf{M})$ is the internal energy function. The smooth function $\chi : \mathcal{B} \rightarrow \mathcal{B}^*$ that assigns to each material point $\mathbf{X} \in \mathcal{B}$ a spatial point $\mathbf{x} \in \mathcal{B}^*$.

In addition, $\mathcal{E}^{\text{elect}}$ in (2.2.14) is the electric energy

$$\mathcal{E}^{\text{elect}} = \frac{\epsilon_0}{2} \int_{\mathcal{B}^*} |\text{grad } \xi|^2 + \int_{\partial \mathcal{B}_d^*} \xi (-\epsilon_0 \text{grad } \xi + \mathbf{p}) \cdot \mathbf{n}, \quad (2.2.16)$$

where $\mathcal{B}^*$ is the domain of the deformed body and $\partial \mathcal{B}_d^*$ is the electric boundary in the current configuration, and $\mathbf{n}$ is the unit normal to the surface $\partial \mathcal{B}_d^*$.

$\mathcal{E}^{\text{mag}}$ in (2.2.14) is the magnetic energy ³

$$\begin{aligned} \mathcal{E}^{\text{mag}} &= \frac{\mu_0}{2} \int_{\mathbb{R}^3} |\text{grad } \zeta|^2 \\ &= \frac{\mu_0}{2} \int_{\mathbb{R}^3} |\text{grad } \zeta^{\text{self}}|^2 - \mu_0 \int_{\mathcal{B}^*} \mathbf{h}^e \cdot \mathbf{m}, \end{aligned} \quad (2.2.17)$$

where μ_0 is the magnetic permeability of free space, $\mathbf{h}^e$ is the far-field applied magnetic field and $\mathbf{m}$ is the magnetization in the current configuration. ζ^{self} is the so-called magnetic self-field which is defined for convenience by decomposing the total magnetic field $\mathbf{h}$ as: $\mathbf{h} = \mathbf{h}_e + \mathbf{h}_{\text{self}}$ and $\mathbf{h}_{\text{self}} = -\text{grad } \zeta^{\text{self}}$ and $\mathbf{h}_{\text{self}} = -\text{grad } \zeta$. Finally, $\mathcal{P}^{\text{mech}}$ in (2.2.14) is defined by

$$\mathcal{P}^{\text{mech}} = - \int_{\partial \mathcal{B}_t} \mathbf{t} \cdot \chi \quad (2.2.18)$$

that is the potential energy related to the dead load $\mathbf{t}$ applied on the traction boundary $\partial \mathcal{B}_t$.

In this work, we will consider the case of a dielectric elastomer subject to two in-plane biaxial forces P_2, P_3 , an applied voltage (2.2.10) and an external far-field magnetic

³The detailed derivation of the form of the magnetic energy and the expression of (2.2.17) can be found in equation (5.1) - (5.5) of the work by Liu.[51]

field (2.2.11) in the thickness direction (see Fig.2.2.1). Considering the assumption of the deformation (see eqs. (2.2.2)-(2.2.7)) and the 1-dimensional nature of the problem, the total free energy of the system shown in Figure 2.2.1, in contrast to the general form (2.2.14) in the three-dimensional space, is given by [6, 48]

$$\begin{aligned} \frac{F}{L_1 L_2 L_3} = & \frac{1}{L_1} \int_0^{L_1} W(\lambda_2, \lambda_3, P, M) dX \\ & + \frac{1}{L_1} \int_0^{l_1} \frac{\epsilon_0}{2} \lambda_2 \lambda_3 (\xi_{,x})^2 dx + \frac{1}{L_1} [\xi \lambda_2 \lambda_3 (-\epsilon_0 \xi_{,x} + p)] \Big|_{x=l_1} \\ & + \frac{1}{L_1} \int_0^{l_1} \frac{\mu_0}{2} \lambda_2 \lambda_3 (\zeta_{,x}^{\text{self}})^2 dx - \frac{1}{L_1} \int_0^{l_1} \lambda_2 \lambda_3 \mu_0 \mathbf{h}^e \cdot \mathbf{m} dx - \frac{P_2 \lambda_2}{L_1 L_3} - \frac{P_3 \lambda_3}{L_1 L_2} \end{aligned} \quad (2.2.19)$$

where the subscript ‘, x ’ denotes the derivative with respect to x in the current configuration.

The free energy expression (2.2.19) contains a mixture of both material and spatial representations. In the following subsection, we will reformulate the free energy expression entirely in the reference configuration.

Material representation of the free energy

Recalling the expression for the stretch λ_1 in (2.2.4), the relationship between the differentials in the current and reference configurations can be written as

$$\xi_{,x} = \xi_{,X} \frac{\partial X}{\partial x} = \frac{\xi_{,X}}{\lambda_1}, \quad \zeta_{,x}^{\text{self}} = \frac{\zeta_{,X}^{\text{self}}}{\lambda_1}, \quad dx = \lambda_1 dX. \quad (2.2.20)$$

In contrast to the spatial forms (2.2.8), the 1D Maxwell equations in the reference configuration are

$$(-\epsilon_0 \lambda_2 \lambda_3 \xi_{,X} + P)_{,X} = 0, \quad (-\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}} + M)_{,X} = 0. \quad (2.2.21)$$

Thus the free energy (2.2.19) can be written in the reference configuration as

$$\begin{aligned}
\frac{F}{L_1 L_2 L_3} = & \frac{1}{L_1} \int_0^{L_1} W dX + \frac{1}{L_1} \int_0^{L_1} \frac{\epsilon_0}{2} (\lambda_2 \lambda_3)^2 (\xi_{,X})^2 dX \\
& + \frac{1}{L_1} \int_0^{L_1} [-\epsilon_0 (\lambda_2 \lambda_3)^2 (\xi_{,X})^2 + \lambda_2 \lambda_3 \xi_{,X} P] dX \\
& + \frac{1}{L_1} \int_0^{L_1} \frac{\mu_0}{2} (\lambda_2 \lambda_3)^2 (\zeta_{,X}^{\text{self}})^2 dX \\
& - \frac{1}{L_1} \int_0^{L_1} \mu_0 h^e M dX - \frac{P_2 \lambda_2}{L_1 L_3} - \frac{P_3 \lambda_3}{L_1 L_2}
\end{aligned} \tag{2.2.22}$$

by using the equations (2.2.20), (2.2.7), and the 1D Maxwell equations (2.2.21) as well as integration by parts.

A vector form of the free energy

There are four generalized coordinates (independent variables) including λ_2 , λ_3 , P and M in the expression of the free energy (2.2.22). Other variables like the electric potential ξ , the magnetic potential ζ and the self-magnetic potential ζ^{self} are related to the four generalized coordinates through the relation (2.2.20) and the Maxwell equations (2.2.21).

We introduce a vector

$$\mathbf{v} = (\lambda_2, \lambda_3, P, M)^T. \tag{2.2.23}$$

Then, a more compact form of the free energy (2.2.22) can be written as:

$$\frac{F(\mathbf{v})}{L_1 L_2 L_3} = \frac{1}{L_1} \int_0^{L_1} W^t(\mathbf{v}) dX - s_2 \lambda_2 - s_3 \lambda_3, \tag{2.2.24}$$

where the total energy density $W^t(\mathbf{v})$ is

$$\begin{aligned}
W^t(\mathbf{v}) = & W(\mathbf{v}) - \frac{\epsilon_0}{2} (\lambda_2 \lambda_3)^2 (\xi_{,X})^2 + \lambda_2 \lambda_3 \xi_{,X} P \\
& + \frac{\mu_0}{2} (\lambda_2 \lambda_3)^2 (\zeta_{,X}^{\text{self}})^2 - \mu_0 h^e M,
\end{aligned} \tag{2.2.25}$$

and the nominal stresses are

$$s_2 = \frac{P_2}{L_1 L_3}, \quad s_3 = \frac{P_3}{L_1 L_2}. \quad (2.2.26)$$

It should be noted that $\xi_{,X}$ and $\zeta_{,X}^{\text{self}}$ in the energy function (2.2.25) are implicitly related to the vector $\mathbf{v}$ in (2.2.23).

2.2.4 Principle of minimum free energy

The equilibrium state $\mathbf{v}$ in (2.2.24) is dictated by the principle of minimum free energy. The free energy should be minimized among all the neighboring states $\mathbf{v} + \delta\mathbf{v}$, $|\delta\mathbf{v}| \ll 1$:

$$F(\mathbf{v}) \leq F(\mathbf{v} + \delta\mathbf{v}). \quad (2.2.27)$$

The inequality (2.2.27) gives the following conditions for the first and second variations:

$$\delta F(\mathbf{v}) = 0 \quad (2.2.28)$$

and

$$\delta^2 F(\mathbf{v}) \geq 0. \quad (2.2.29)$$

Using (2.2.28) and (2.2.29), the first variation condition can be written as:

$$\frac{\delta F}{L_1 L_2 L_3} = \frac{1}{L_1} \int_0^{L_1} \delta W^t(\mathbf{v}) dX - s_2 \delta \lambda_2 - s_3 \delta \lambda_3 = 0 \quad (2.2.30)$$

while the second variation condition is an integral inequality, such that

$$\frac{\delta^2 F}{L_1 L_2 L_3} = \frac{1}{L_1} \int_0^{L_1} \delta^2 W^t(\mathbf{v}) dX \geq 0. \quad (2.2.31)$$

We remark that the first and second variations are integral quantities since we have allowed for inhomogeneous states. For homogeneous deformation and perturbation, a direct use of the Hessian matrix approach is often used to study stability [15, 40]. Detailed derivations of the first and second variations can be found in the Appendix.

2.3 Homogeneous deformation of ideal dielectric elastomer

2.3.1 Homogeneous deformation

In order to make the calculation simpler and obtain clear insights regarding our stated objective, we now limit our attention to the homogeneous deformation. That is, the deformation gradient is constant everywhere in the deformed body. This assumption has been used quite frequently to study electromechanical instability [15, 13, 40].

For homogeneous deformation of incompressible materials, the deformation gradient (2.2.3) reduces to

$$\mathbf{F} = \begin{pmatrix} 1/\lambda_2\lambda_3 & 0 & 0 \\ 0 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{pmatrix}, \quad (2.3.1)$$

where λ_1, λ_2 , and λ_3 are undetermined constants that are independent of the coordinates.

In addition, the free energy (2.2.24) reduces to

$$\frac{F}{L_1 L_2 L_3} = W^t(\mathbf{v}) - s_2 \lambda_2 - s_3 \lambda_3, \quad (2.3.2)$$

the first variation condition (2.2.30) becomes

$$\frac{\delta F}{L_1 L_2 L_3} = \frac{\partial W^t}{\partial \mathbf{v}} \cdot \delta \mathbf{v} - s_2 \delta \lambda_2 - s_3 \delta \lambda_3 = 0 \quad (2.3.3)$$

and the second variation condition (2.2.31) reads

$$\frac{\delta^2 F}{L_1 L_2 L_3} = \delta^2 W^t(\mathbf{v}) = \delta \mathbf{v} \cdot \frac{\partial^2 W^t}{\partial \mathbf{v}^2} \delta \mathbf{v} = \delta \mathbf{v} \cdot \mathbb{H} \delta \mathbf{v} \geq 0. \quad (2.3.4)$$

This stability condition $\delta^2 F \geq 0$ only requires a positive-definite (Hessian) matrix $\mathbb{H} = \frac{\partial^2 W^t}{\partial \mathbf{v}^2}$ at equilibrium.

2.3.2 Ideal dielectric elastomer

The formulation in the preceding sub-sections is applicable for any soft dielectric material with the internal energy $W(\mathbf{v})$. To produce specific results, we will make a choice of constitutive law and consider an ideal dielectric material [6, 51, 48]

$$W = \frac{c}{2}(\lambda_2^2 + \lambda_3^2 + \lambda_2^{-2}\lambda_3^{-2} - 3) + \frac{P^2}{2\epsilon_0(\hat{\epsilon}_r - 1)} + \frac{\mu_0 M^2}{2(\hat{\mu}_r - 1)}, \quad (2.3.5)$$

where c is the small-strain shear modulus, $\hat{\epsilon}_r$ and $\hat{\mu}_r$ are the relative electric permittivity and magnetic permeability of the film, respectively.

The first term in (2.3.5) denotes the mechanical part, while the second and third terms are the electrical and magnetic parts of the internal energy, respectively. In the absence of electric and magnetic fields, the internal energy simply represents an incompressible neo-Hookean elastic material.

2.3.3 Equilibrium solutions

With (6.0.7) and (2.3.3), the Euler-Lagrange equations are

$$\frac{\partial W^t}{\partial \lambda_2} - s_2 = 0, \quad \frac{\partial W^t}{\partial \lambda_3} - s_3 = 0, \quad \frac{\partial W^t}{\partial P} = 0, \quad \frac{\partial W^t}{\partial M} = 0. \quad (2.3.6)$$

Together with the energy function (2.3.5), we have

$$c(\lambda_2 - \lambda_2^{-3}\lambda_3^{-2}) - \epsilon_0\lambda_2\lambda_3^2(\xi_{,X})^2 + \lambda_3\xi_{,X}P + \mu_0\lambda_2\lambda_3^2(\zeta_{,X}^{\text{self}})^2 - s_2 = 0, \quad (2.3.7a)$$

$$c(\lambda_3 - \lambda_2^{-2}\lambda_3^{-3}) - \epsilon_0\lambda_2^2\lambda_3(\xi_{,X})^2 + \lambda_2\xi_{,X}P + \mu_0\lambda_2^2\lambda_3(\zeta_{,X}^{\text{self}})^2 - s_3 = 0, \quad (2.3.7b)$$

$$\frac{P}{\epsilon_0(\hat{\epsilon}_r - 1)} + \lambda_2\lambda_3\xi_{,X} = 0, \quad \text{and} \quad (2.3.7c)$$

$$\frac{\mu_0 M}{(\hat{\mu}_r - 1)} + \mu_0(\lambda_2\lambda_3)^2\zeta_{,X}^{\text{self}}[\zeta_{,X}^{\text{self}}]_{,M} - \mu_0 h^e = 0. \quad (2.3.7d)$$

In this set of four algebraic equations, we have four unknown independent variables, λ_2 , λ_3 , P and M . The remaining two variables $\xi_{,X}$ and $\zeta_{,X}^{\text{self}}$ are related to these four independent variables through the relation (2.2.20) and the Maxwell equations (2.2.21). Together with the electric and magnetic boundary conditions, we can solve this set of algebraic equations.

In the following, we give the details of the solution.

Solution of the polarization and the electric field

From (2.3.7c), we can easily obtain the polarization P in the reference configuration as

$$P = -\epsilon_0(\widehat{\epsilon}_r - 1)\lambda_2\lambda_3\xi_{,X}. \quad (2.3.8)$$

With (2.2.7), (2.2.9) and (2.2.20), the polarization can be written in the current configuration as

$$p = -\epsilon_0(\widehat{\epsilon}_r - 1)\xi_{,x}, \quad (2.3.9)$$

which is consistent with the constitutive relation stated earlier in this work.

Substituting (2.3.9) into the Maxwell equation (2.2.8), we can obtain a Laplace equation of the electric potential ξ . Together with the electric boundary condition (2.2.10), the solution of the potential in the current configuration is given by

$$\xi = \frac{V}{l_1}x, \quad 0 \leq x \leq l_1. \quad (2.3.10)$$

Then the electric fields in the current and reference configurations are

$$-\xi_{,x} = -\frac{V}{l_1} \quad (2.3.11)$$

and

$$-\xi_{,X} = -\lambda_1\xi_{,x} = -\frac{V}{L_1}. \quad (2.3.12)$$

For further discussion, we define the magnitude of the nominal electric field as:

$$\tilde{E} = \frac{V}{L_1}. \quad (2.3.13)$$

Solution of the magnetization and the magnetic field

Similar to the polarization, from (2.3.7d), we can get the magnetization M in the reference configuration as

$$M = (\hat{\mu}_r - 1)(-\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}} + h^e). \quad (2.3.14)$$

With (2.2.7), (2.2.9) and (2.2.20), the magnetization in the current configuration is

$$m = (\hat{\mu}_r - 1)(-\zeta_{,x}^{\text{self}} + h^e) = -(\hat{\mu}_r - 1)\zeta_{,x}, \quad (2.3.15)$$

which, as expected, agrees with our constitutive relation.

Substituting (2.3.15) into the Maxwell equations (2.2.8), we have a Laplace equation of the potential ζ . Together with the magnetic boundary conditions (2.2.11), (2.2.12) and (2.2.13), the solution of the magnetic field in the current configuration is given by

$$-\zeta_{,x} = \begin{cases} \frac{1}{\hat{\mu}_r} h^e, & 0 < x < l_1, \\ h^e, & \text{otherwise.} \end{cases} \quad (2.3.16)$$

Then the magnetization and the self-magnetic field in the current configuration are given by

$$m = \begin{cases} \tilde{h}, & 0 < x < l_1, \\ \hat{\mu}_r \tilde{h}, & \text{otherwise,} \end{cases} \quad (2.3.17)$$

and

$$-\zeta_{,x}^{\text{self}} = \begin{cases} -\tilde{h}, & 0 < x < l_1, \\ 0, & \text{otherwise,} \end{cases} \quad (2.3.18)$$

where

$$\tilde{h} = \frac{(\hat{\mu}_r - 1)}{\hat{\mu}_r} h^e. \quad (2.3.19)$$

Using the relation (2.2.20), the magnetic field, the magnetization and the self-magnetic

field in the reference configuration are

$$-\zeta_{,X} = \frac{-\zeta_{,x}}{\lambda_2 \lambda_3}, \quad M = m \quad \text{and} \quad -\zeta_{,X}^{\text{self}} = \frac{-\zeta_{,x}^{\text{self}}}{\lambda_2 \lambda_3}. \quad (2.3.20)$$

Solution for the stretches

By substituting (2.3.8), (2.3.12) and (2.3.20) into (2.3.7a) and (2.3.7b), we have

$$c(\lambda_2 - \lambda_2^{-3} \lambda_3^{-2}) - \epsilon_0 \hat{\epsilon}_r \lambda_2 \lambda_3^2 \tilde{E}^2 + \mu_0 \lambda_2^{-1} \tilde{h}^2 - s_2 = 0 \quad \text{and} \quad (2.3.21)$$

$$c(\lambda_3 - \lambda_2^{-2} \lambda_3^{-3}) - \epsilon_0 \hat{\epsilon}_r \lambda_2^2 \lambda_3 \tilde{E}^2 + \mu_0 \lambda_3^{-1} \tilde{h}^2 - s_3 = 0. \quad (2.3.22)$$

For the sake of convenience, we introduce the following dimensionless variables by appropriate normalization

$$\bar{s}_2 = \frac{s_2}{c}, \quad \bar{s}_3 = \frac{s_3}{c}, \quad E = \tilde{E} \sqrt{\frac{\epsilon_0 \hat{\epsilon}_r}{c}} \quad \text{and} \quad H = \tilde{h} \sqrt{\frac{\mu_0}{c}}. \quad (2.3.23)$$

Then (2.3.21) and (2.3.22) can be recast as

$$(\lambda_2 - \lambda_2^{-3} \lambda_3^{-2}) - \lambda_2 \lambda_3^2 E^2 + \lambda_2^{-1} H^2 - \bar{s}_2 = 0 \quad \text{and} \quad (2.3.24)$$

$$(\lambda_3 - \lambda_2^{-2} \lambda_3^{-3}) - \lambda_2^2 \lambda_3 E^2 + \lambda_3^{-1} H^2 - \bar{s}_3 = 0. \quad (2.3.25)$$

For given values of control parameters (the dead loads $\bar{s}_2$, $\bar{s}_3$, the electric field E and the magnetic field H) defined in (2.3.23), the two equilibrium equations (2.3.24) and (2.3.25) determine the values of the two stretches λ_2 and λ_3 .

2.3.4 Stability analysis

According to principle of minimum energy (2.2.27), a homogeneously deformed dielectric will be stable under small perturbations in control parameters when the Hessian matrix $\mathbb{H}$ in (6.0.12) is positive-definite. By substituting the energy function (2.3.5) into the Hessian matrix (6.0.12), we obtain:

$$\mathbb{H} = \begin{pmatrix} \mathbb{H}_{11} & \mathbb{H}_{12} & \mathbb{H}_{13} & \mathbb{H}_{14} \\ & \mathbb{H}_{22} & \mathbb{H}_{23} & \mathbb{H}_{24} \\ & & \mathbb{H}_{33} & \mathbb{H}_{34} \\ & \text{Sym} & & \mathbb{H}_{44} \end{pmatrix}, \quad (2.3.26)$$

where the entries, at the equilibrium solutions (2.3.8), (2.3.12), (2.3.14), (2.3.18)-(2.3.25), are

$$\mathbb{H}_{11} = c(1 + 3\lambda_2^{-4}\lambda_3^{-2}) - \epsilon_0\lambda_3^2\tilde{E}^2 + \mu_0\lambda_2^{-2}\tilde{h}^2, \quad (2.3.27a)$$

$$\mathbb{H}_{12} = 2c\lambda_2^{-3}\lambda_3^{-3} - \epsilon_0(\hat{e}_r + 1)\lambda_2\lambda_3\tilde{E}^2 + 2\mu_0\lambda_2^{-1}\lambda_3^{-1}\tilde{h}^2, \quad (2.3.27b)$$

$$\mathbb{H}_{13} = \lambda_3\tilde{E}, \quad (2.3.27c)$$

$$\mathbb{H}_{14} = 2\mu_0\lambda_2^{-1}\tilde{h}, \quad (2.3.27d)$$

$$\mathbb{H}_{22} = c(1 + 3\lambda_2^{-2}\lambda_3^{-4}) - \epsilon_0\lambda_2^2\tilde{E}^2 + \mu_0\lambda_3^{-2}\tilde{h}^2, \quad (2.3.27e)$$

$$\mathbb{H}_{23} = \lambda_2\tilde{E}, \quad (2.3.27f)$$

$$\mathbb{H}_{24} = 2\mu_0\lambda_3^{-1}\tilde{h}, \quad (2.3.27g)$$

$$\mathbb{H}_{33} = \frac{1}{\epsilon_0(\hat{e}_r - 1)}, \quad (2.3.27h)$$

$$\mathbb{H}_{34} = 0, \quad \text{and} \quad (2.3.27i)$$

$$\mathbb{H}_{44} = \frac{\mu_0\hat{\mu}_r}{\hat{\mu}_r - 1}. \quad (2.3.27j)$$

In what follows we will limit our discussion to the determinant of the Hessian matrix rather than all its principal minors. For prescribed dead loads P_2 and P_3 and external magnetic field h^e , for example, changing the voltage V takes the system from a state of stable equilibrium to a critical state specified by the condition:

$$\det(\mathbb{H}) = 0. \quad (2.3.28)$$

Beyond that, the determinant of the Hessian matrix becomes negative and the equilibrium states are no longer stable.

2.4 Stability and emergent magnetoelectricity

2.4.1 In the absence of external magnetic field

To connect with past work on dielectric elastomers, we first consider the scenario when magnetic fields are absent. For $H = 0$, the equilibrium equations (2.3.24) and (2.3.25) reduce to

$$(\lambda_2 - \lambda_2^{-3}\lambda_3^{-2}) - \lambda_2\lambda_3^2E^2 - \bar{s}_2 = 0 \quad \text{and} \quad (2.4.1)$$

$$(\lambda_3 - \lambda_2^{-2}\lambda_3^{-3}) - \lambda_2^2\lambda_3E^2 - \bar{s}_3 = 0. \quad (2.4.2)$$

These two equations are equivalent to eqs.(6a) and (6b) in the work of Zhao and Suo [15] with an appropriate change of notation. We remark that the state variables used in the work [15] are the stretch and the nominal electric displacement $\tilde{D}$ while we have chosen to use the stretch and the nominal polarization P . The connection between various flavors of electromagnetic theories of deformable media has been discussed at length by Liu [51].

2.4.2 Effect of the magnetic field on the equilibrium

In this section, we investigate the effect of an external magnetic field. For simplicity of presentation, we consider the special case of equal biaxial stresses, such that $\bar{s}_2 = \bar{s}_3 = S$ in the equilibrium equations (2.3.24) and (2.3.25). As a result, $\lambda_2 = \lambda_3 = \lambda$ and the two equilibrium equations become:

$$(\lambda - \lambda^{-5}) - \lambda^3E^2 + \lambda^{-1}H^2 - S = 0, \quad (2.4.3)$$

where the dimensionless electric and magnetic fields E and H are defined by (2.3.23). The dimensionless magnetic field H is related to the magnetic parameter $\tilde{h}$ in (2.3.19). It is important to note that the magnetic permeability should be $\mu_r = \hat{\mu}_r\mu_0 > \mu_0$ for the emergent magnetoelectric effect to manifest. Therefore, $\tilde{h} = (\hat{\mu}_r - 1)h^e/\hat{\mu}_r > 0$ and $H > 0$ are chosen in our numerical plots of the equilibrium (2.4.3) and the stability (2.3.26). Note that S, E

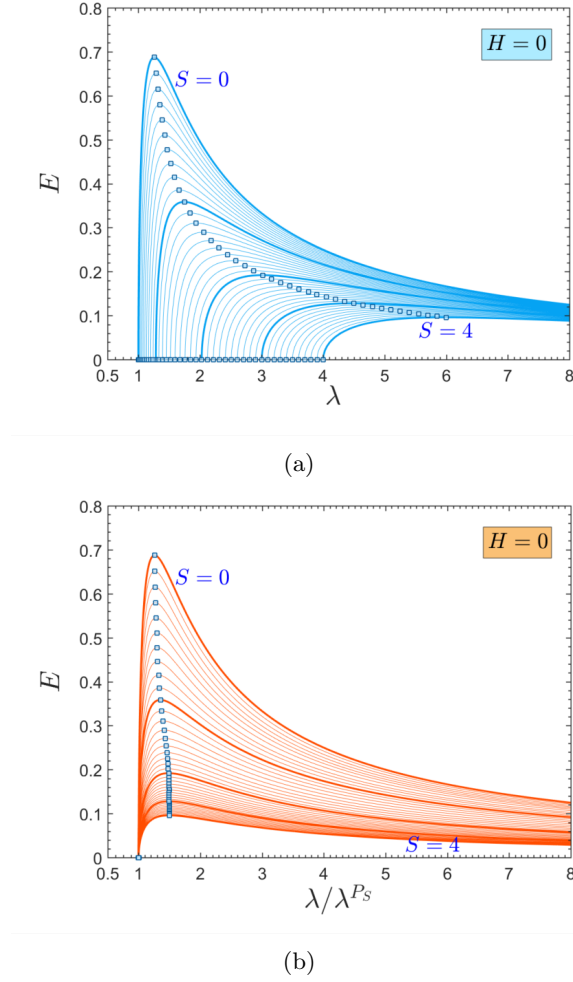


Figure 2.4.1: Behavior of a neo-Hookean dielectric film subjected to a zero magnetic field under various equal-biaxial loads from $S = 0$ to $S = 4$ in equilibrium: (a) in-plane stretch λ *vs.* E , (b) in-plane actuation stretch λ/λ^{Ps} *vs.* E .

and H are control parameters that result in the stretch λ . In the absence of the magnetic field, $H = 0$, (2.4.3) will yield eq.(8)₂ in the work of Zhao and Suo [15].

For the material properties suggested in the reference [15], the shear modulus of soft dielectric is $c = 10^6$ N/m², the material permittivity is $\varepsilon_0 \hat{\varepsilon}_r = 4 \times 10^{-11}$ F/m, and the nominal electric field is in the order $\tilde{E} = 10^8$ V/m in (2.3.23). Then the dimensionless electric E in (2.3.23) is equal to $E = 0.63$. For a dimensionless magnetic field $H = 0.5$ in (2.3.23), we need to input an external magnetic field $\tilde{h} = 0.55$ T (tesla), $\tilde{h} \approx h^e$ for large relative magnetic permeability $\hat{\mu}_r$ in (2.3.19), by using the vacuum permeability is

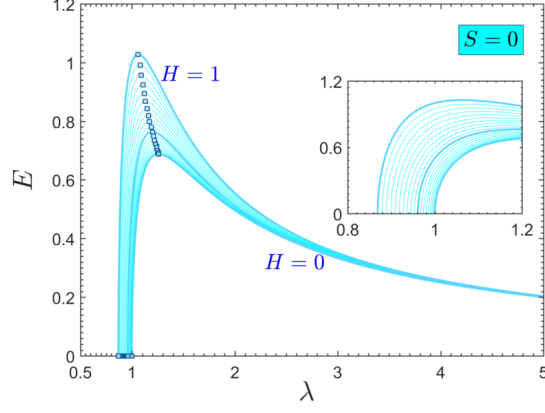
$\mu_0 = 4\pi \times 10^{-7} \text{ T} \cdot \text{m/A}$. This external magnetic field can decrease the true electric field from $1.25 \times 10^8 \text{ V/m}$ ($E = 0.63$ and $H = 0$) to $1.07 \times 10^8 \text{ V/m}$ ($E = 0.63$ and $H = 0.5$), which can avoid the electric breakdown of soft dielectrics in some circumstances.

In Fig. 2.4.1, we show the stretch λ and the actuation stretch λ/λ^{Ps} of the dielectric film subject to an applied voltage and various dead loads in the absence of a magnetic field. For simplicity, only the case of non-negative E in (2.4.3) is plotted. Fig. 2.1(a) shows that the nominal electric field E increases nonmonotonically with the increase of the in-plane stretch λ under an equal-biaxial load S . Alternatively, the electric field and the dead load can increase the in-plane stretch, that is, they can expand the dielectric film. At a zero electric field $E = 0$, the higher the dead load S , the larger the stretch λ .

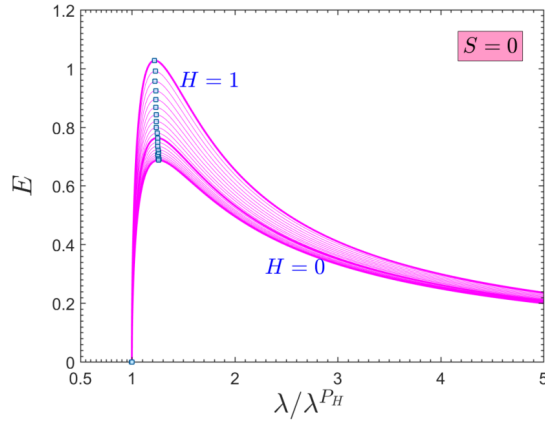
For every value of the dead load S , there exists a peak that corresponds to the maximum of the nominal electric field in equilibrium. The maximum E in each curve decreases with the increase of the dead load S and the peak moves from a low stretch (the left) to a high stretch (the right). A point in each curve corresponds to an equilibrium state whose stability can be verified by the positive-definiteness of the Hessian matrix (2.3.26). The peak of each curve corresponds to $\det(\mathbb{H}) = 0$, each point on the left-hand side corresponds to a positive-definite Hessian (stable), each point on the right-hand side corresponds to a non-positive-definite Hessian (unstable).

Fig. 2.1(b) shows how the nominal electric field E affects the actuation stretch. The actuation stretch here is defined as the ratio of the stretch λ to the prestretch λ^{Ps} that is only induced by the dead load. Each curve in Fig. 2.1(b) starts at the unit actuation stretch and a zero electric field, and then the trend and the stability of each curve is similar to that in Fig. 2.1(a). These results are to be expected and simply presented as benchmark. For detailed discussion of these non-magnetic behaviors, the reader can refer to the work [15, 13, 40].

In Fig. 2.4.2, we present the stretch λ and the actuation stretch λ/λ^{Ph} of the dielectric



(a)



(b)

Figure 2.4.2: Behavior of a neo-Hookean dielectric film for $S = 0$ under various magnetic fields from $H = 0$ to $H = 1$ in equilibrium: (a) in-plane stretch λ vs. E , (b) in-plane actuation stretch λ/λ^{P_H} vs. E .

film subject to an applied voltage and various magnetic fields. For simplicity, all these curves are plotted at a zero dead load $S = 0$. In Fig. 2.2(a), with the increase of the magnetic field from $H = 0$ to 1, the peak in each curve increases significantly and moves slightly from a high stretch (the right) a low stretch (the left). These changes indicate that the magnetic field *increases* the critical electrical field (the electric field at the peak of each curve). However, the shift of the peak (from right to left) due to the increase of H in Fig. 2.2(a) is opposite to that observed in Fig. 2.1(a) (from left to right) resulting from the increase in S . This indicates that the magnetic field squeezes the dielectric film and then decreases the in-plane stretch. Indeed, the magnetic field H has an opposite effect on the

in-plane stretch λ compared to the electric field E and the dead load S . The magnetic field compresses the film in-plane while the electric field and the dead load expands it. At a zero electric field $E = 0$, for example, increasing the magnetic field H changes the stretch λ from 1 (at $H = 0$) to 0.87 (at $H = 1$).

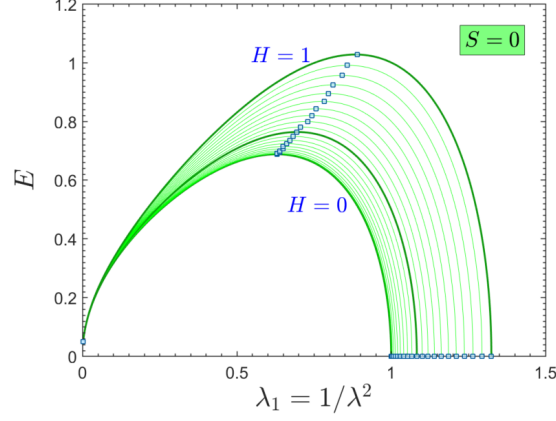
The prestretch λ^{P_H} , in contrast to λ^{P_S} in Fig.2.1(b), is defined as the stretch induced solely by the magnetic field. Then the actuation stretch by the electric field here is defined as the ratio λ/λ^{P_H} . Unlike the maximum electric field, the actuation stretch of the peak on each curve is insensitive to the increase of the magnetic field.

From the previous discussion of Fig. 2.4.1, we know that the magnetic field squeezes the dielectric film in-plane. Since the film is incompressible, the thickness of the dielectric film will increase.

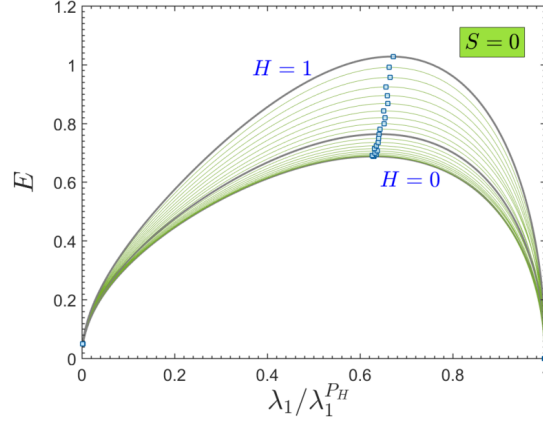
Fig. 2.4.3 shows the stretch $\lambda_1 = 1/\lambda^2$ and the actuation stretch $\lambda_1/\lambda_1^{P_H}$ in the thickness direction of the film. To make a direct comparison between the electric and magnetic fields, we choose a zero dead load here. In Fig. 2.3(a), at a zero electric field $E = 0$, the magnetic field H can increase the thickness from an initial stretch of 1 (at $H = 0$) to a stretch of 1.32 (at $H = 1$). With the increase of the electric field, there exists an apparent competition between the electric and magnetic fields. At a low electric field but a relatively high magnetic field, the stretch is greater than one (increasing thickness); at a high electric field but a relatively low magnetic field, the stretch is less than one (decreasing thickness). It is worth mentioning that each point in Fig. 2.4.3 on the left-hand side of the peak corresponds to a non-positive-definite Hessian (unstable) while each point on the right-hand side corresponds to a positive-definite Hessian (stable), unlike Figs. 2.4.1 and 2.4.2.

In Fig. 2.3(b), the actuation stretch in the thickness direction is plotted. Note that the actuation stretch induced by the electric field compresses the film in the thickness direction. Thus the actuation stretch is always less than 1 in Fig. 2.3(b) but greater than 1 in Fig. 2.2(b).

In Figs. 2.4.1, 2.4.2 and 2.4.3, we examine the effect of a varying nominal electric field (the vertical axis) while fixing the external magnetic field and the dead load on the



(a)



(b)

Figure 2.4.3: Behavior of a neo-Hookean dielectric film for $S = 0$ under various magnetic fields from $H = 0$ to $H = 1$ in equilibrium: (a) in-thickness stretch $\lambda_1 = 1/\lambda^2$ vs. E , (b) in-thickness actuation stretch $\lambda_1/\lambda_1^{P_H}$ vs. E .

in-plane and in-thickness stretches (the horizontal axis). The electric and magnetic fields have opposite effects on the in-plane and in-thickness stretches. To further investigate the role of magnetic field, we plot the variation of the in-plane stretch as a result of a varying magnetic field (the vertical axis) at constant nominal electric field and dead load. The results are shown in Fig. 2.4.4 for a zero dead load for simplicity. We first note that the plot is symmetric about the horizontal axis. This is expected mathematically because the magnetic field appears a squared term in the equilibrium equation (2.4.3) which makes it independent of the sign. From a physical point of view, the Maxwell stress is a quadratic form of the magnetic field, and thus the equilibrium state is the same regardless of the magnetic field

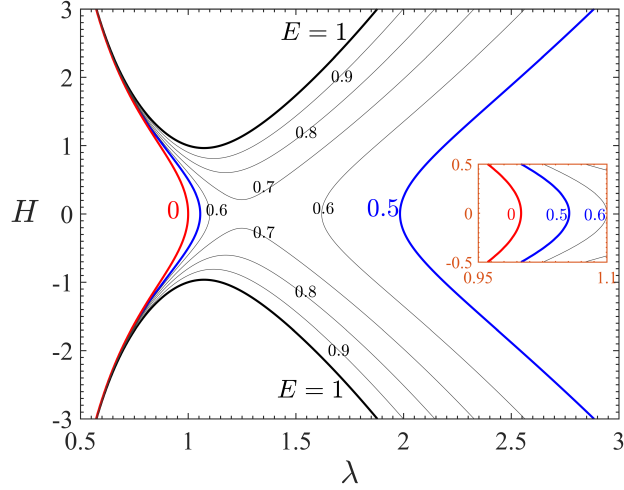


Figure 2.4.4: Behavior of a neo-Hookean dielectric film at a zero dead load $S = 0$ under various nominal electric field from $E = 0$ to $E = 1$ in equilibrium: in-plane stretch λ *vs.* magnetic field H .

direction. In addition, for a prescribed nominal electric field E , the equilibrium equation (2.4.3) yields two curves. For an E less than the critical value, between 0.6 and 0.7, the two equilibrium curves are separated on the left and on the right. On the other hand, for an E greater than the critical E , the two equilibrium curves are separated on the top and bottom. In each curve, there exists a turning point. For the up (down) curves, the turning point denotes a minimum (maximum) of the magnetic field; for the left (right) curves, it denotes the maximum (minimum) of the in-plane stretch. Note that a point in each curve corresponds to an equilibrium state and the stability can be verified by the positive definite of the Hessian matrix.

In what follows, we will show that if a point on each curve is on the left-hand side of the turning point, the Hessian is positive-definite and the point stands for a stable equilibrium state. On the contrary, if the point is on the right-hand side of the turning point, it denotes an unstable equilibrium state. Therefore, the curve on the left-hand is always stable while the curve on the right-hand side is unstable. For up and down curves, the parts on the left-hand of the turning points are stable, and the right part is unstable.

2.4.3 Effects of the magnetic field on stability

Each curve in Fig. 2.4.1, 2.4.2, and 2.4.3 reveals a maximum which corresponds to $\det(\mathbb{H}) = 0$. To further illustrate the magneto-electro-mechanical stability, we consider the sign of the determinant $\det(\mathbb{H})$ of an uniaxial stretched thin film subject to electric and magnetic fields in equilibrium.

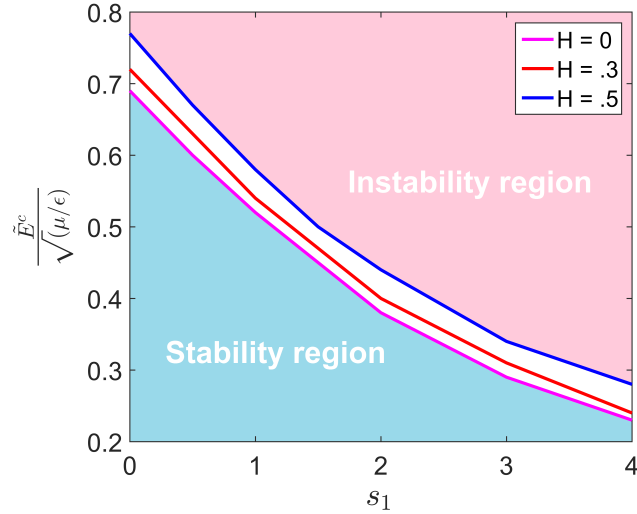


Figure 2.4.5: Stability and instability regions in the dead load (s_1) - electric field plane of a neo-Hookean dielectric film under various magnetic fields $H = 0, 0.3, 0.5$.

Fig. 2.4.5 shows the regions of stability and instability for a film of soft materials subject to a uniaxial tension s_1 and a constant electric field under various external magnetic fields. We plot three curves and each of them corresponds to the zero determinant $\det(\mathbb{H}) = 0$ at a given value of the magnetic field, $H = 0, 0.3, 0.5$. For a given H , the curve in Figure 2.4.5 represents the variation of the critical electric field $\tilde{E}^c$ at which $\det(\mathbb{H}) = 0$ with respect to the applied dead load. The determinant is positive (stable) below the curve while it is negative (unstable) above the curve.

It is clear from Fig. 2.4.5 that the external magnetic field enhances the magneto-electro-mechanical stability. The curve with a higher magnetic field H is always above the curve with a lower H . Without considering the magnetic field $H = 0$, the curve would be the

lowest. A higher curve means a larger stability region that is enclosed by the curve of zero determinant and the axes. This clearly shows that the magnetic field allows the film to sustain a higher electric field. We remark that the electric breakdown is not taken into account here.

2.5 Wireless actuation and energy harvesting

A key insight evident from the discussion in the preceding paragraphs is that the presence of external magnetic field increases the critical nominal electric field and reduces the critical actuation stretch, thus suppressing pull-in instability. The critical value of the nominal electric field corresponds to the intersection point of the three stresses acting on the dielectric material: the mechanical stress, the electric Maxwell stress and the magnetic Maxwell stress. At a constant external magnetic field, the latter attempts to “squeeze” the material, thus reducing its actuation stretch and increasing its thickness. Changing the external voltage while maintaining constant external magnetic field and mechanical stress will affect the nominal electric field, thus changing the electric Maxwell stress. Since magnetic stress acts against the electric and the mechanical stresses, the material is able to withstand a larger critical electric field, but the critical actuation stretch will be smaller and depends on the magnitude of the magnetic field. Beyond that critical point, any small perturbation will move the film to an unstable state where it fails without reaching an equilibrium. The same effect can be seen in the case of a uniaxial stress (see Fig. 2.4.5); the presence of the magnetic field also reduces the actuation stretch and increases the critical electric field as can be seen in Fig. 2.4.2 and 2.4.3.

We have shown that the applied electric voltage and the external magnetic field have opposite effects on the deformation of a soft dielectric, that is, the voltage makes the film thinner while the magnetic field makes the film thicker. Based on these results, along with the concept of a simple electric capacitor where capacitance decreases as thickness increases, we propose a simple design to increase the voltage between an isolated charged film by applying an external magnetic field. A higher voltage can be exploited to do useful work—shown

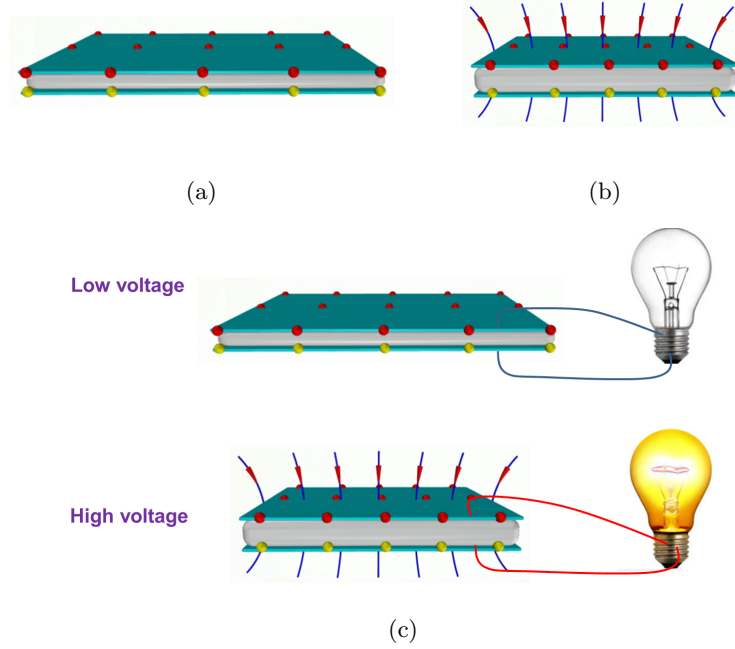


Figure 2.5.1: Fixed total charges on the top and bottom layers of a dielectric film: (a) No magnetic field, (b) With external magnetic field. (c) A higher voltage can power a connected device.

schematically in form of powering a light-bulb (see Fig. 2.5.1). To get a rough estimate of how much energy can be harnessed in this case, we consider the simple model of a dielectric thin film with dimensions $L_1 = 10$ mm, $L_2 = 10$ mm and $L_3 = 1$ mm. We consider the shear modulus of the soft material $c = 10^6$ N/m², the electric permittivity $\epsilon_0 \hat{\epsilon}_r = 4 \times 10^{-11}$ F/m and magnetic permittivity $\hat{\mu}_r = 5$. Then, we fix surface charge of density $q_0 = 1 \times 10^{-3}$ C/m² to the upper surface and $-q_0$ to the lower surface of the thin film. Before applying the magnetic field, the capacitance of this dielectric capacitor is $C = \frac{Q}{V_0} = \frac{\epsilon_0 \hat{\epsilon}_r L_1 L_2}{L_3}$. Note that the total charge on each surface, $Q = q_0 L_1 L_2$, is also conserved. Thus, the electric potential energy stored in it is $U_0 = \frac{1}{2} \frac{Q^2}{C} = \frac{1}{2} \frac{Q^2}{\epsilon_0 \hat{\epsilon}_r} \frac{L_3}{L_1 L_2} \approx 1.25 \times 10^{-3}$ J. Normally, a magnetic field of 0.5 T (equivalent to 0.5×10^4 Oe) can deform the thin film and increase its thickness by about 10%. Substitute this 10% change of the thickness L_3 , then we know that the electric potential energy stored in the capacitor can be increased by more than 20%. This amount of potential energy ($\sim 0.2U_0 = 250\mu\text{J}$) comes from the magnetic field. If the frequency of the magnetic field is 20 Hz, then the output power due to the magnetic field is

$\sim 250\mu J \times 20s^{-1} = 5 \text{ mW}$. This power is enough to power a mini single die LED. We need to note that this is a very small piece of thin film. In reality, we can further enhance the output power by increasing the size of the thin film or even changing the magnetic permittivity of the material. In summary, the external magnetic field increases the voltage on the film and can be used for wireless energy harvesting.

2.6 Concluding remarks

In this chapter, we have explored the magneto-electro-mechanical behavior and instability of soft materials under the combined action of mechanical, electrical and magnetic loads. As long as the magnetic permeability of the soft matter is larger than that of vacuum, an emergent magnetoelectric effect appears due to the interaction of deformation and a pre-existing electric field. While this insight has been appreciated before [6], our key emphasis is to explore the instability behavior of such a system. Our formulation is relatively general although, for illustrative results, we primarily focus on thin films and homogeneous deformation of an ideal neo-Hookean elastomer. Even for this simple case, the insights are rich. The presence of an external magnetic field gives us an important control variable to impact the equilibrium behavior of the dielectric thin film. In particular, pull-in instability can be significantly suppressed by applying an external magnetic field. As a result, the stability of the dielectric film is enhanced which allows it to sustain larger electric fields and mechanical loads. In contrast to the conventional interplay between mechanical and electrical fields, the interaction of three fields provides interesting opportunities to harness large deformation and instabilities of soft dielectrics and presents tantalizing prospects for wireless energy harvesting. Our work provides a simple basis to further explore magnetoelectric wireless energy harvesting devices. Further research on magneto-electro-mechanical instabilities could be elaborated to investigate the post-bifurcation analysis and the effects of the magnetic field on wrinkling, creasing, and cratering as well as other types of instabilities.

Chapter 3

Using Electrets to Design Concurrent Magnetoelectricity and Piezoelectricity in Soft Materials

Piezoelectricity and magnetoelectricity are contradictory properties with a rather limited set of natural (often hard) materials that exhibit both. Composite materials, almost always restricted to hard ones, provide a limited recourse with the attendant limitations of small strains, fabrication challenges among others. In this article, using the concept of electrets, we propose a simple scheme to design soft, highly deformable materials that simultaneously exhibit piezoelectricity and magnetoelectricity. We demonstrate that merely by embedding charges and ensuring elastic heterogeneity, the geometrically nonlinear behavior of soft materials leads to an emergent piezoelectric and magnetoelectric behavior. We find that, an electret configuration made of sufficiently soft (non-piezoelectric and non-magnetic) polymer foams can exhibit simultaneous magnetoelectricity and piezoelectricity with large coupling constants that exceed the best-known ceramic composites. Moreover, we show that these properties can be tuned with the action of an external field. Expanding this work to study more complex configurations could pave the way to optimizing multifunctional materials and their promising future applications.

3.1 Introduction

The ability to control magnetization by an external electric field or polarization by a magnetic field has promising technological applications. These include, among many others, spintronics, non-volatile memories [47], wireless energy transfer [46], multiple-state memory bits, and memories that can be written electrically and accessed magnetically [43]. Similarly, piezoelectricity, where a uniform mechanical strain can induce an electric field and conversely, a uniform electric field can cause mechanical actuation, has also found wide applications e.g. energy harvesting, artificial muscles, sensing and actuation, advanced microscopy, minimally invasive surgery among others [53, 54, 3].

Historically, the magnetoelectric effect was predicted as early as 1894 while magnetoelectric materials were not discovered until the 1950's. Since then, extensive research has gone into the quest for a strong magnetoelectric (ME) effect. Disappointingly, the ME effect is intrinsic only in a few single phase materials and the coupling parameter itself ($1 \sim 20 mV/cmOe$) is too small to be used effectively in several device realizations. Moreover, most of these materials have Curie or Neel temperature far below the room temperature. Growth of good crystals also represented a major challenge since it calls for expensive materials and processing techniques [1]. As a result, the interest in ME effect declined, and all the applications which were foreseen at the time of its discovery were regarded as unpromising [2].

The revival of this research field came in the 1990's where the focus shifted on engineering magnetoelectricity through composite materials. An artificial way of designing magnetoelectric materials may be realized using the concept of product properties [44]; that is, coupling magnetic and electric degrees of freedom indirectly through a third parameter such as mechanical deformation. Combining piezomagnetic and piezoelectric phases or magnetostrictive and piezoelectric phases can yield the desired property. Typically, in these composites, an external magnetic field creates mechanical strain in the magnetostrictive material, which is transferred to the adjacent piezoelectric material. This, in turn, generates an electric field due to the stress generated in the piezoelectric component of the

composite. This way, magnetoelectricity can be engineered in materials which intrinsically do not allow the ME effect [2].

While the field of piezoelectrics is more mature, most materials exhibiting a high electromechanical coupling are hard brittle ceramics such as perovskite ferroelectrics. Soft magnetoelectric materials are virtually non-existent and the few soft piezoelectric materials (e.g. Polyvinylidene difluoride) are only weakly piezoelectric (as compared to crystalline, hard and brittle ferroelectrics). Multifunctional soft materials, with their attendant advantages of cost-effectiveness, ease of fabrication and suitability for large deformation, can pave the way for a suite of applications e.g., soft robotics and machines, flexible and stretchable electronics among others [55, 56, 57].

While the phenomenon of magnetoelectricity alone is rather elusive, engineering the combined presence of both piezoelectricity and magnetoelectricity in soft materials, in amounts that are suitable for device applications, is a tall order.

In this work, based on our recent work on the concept of electrets in soft materials [6, 9], we propose a mechanism that can be used to simultaneously create both a large (apparent) magnetoelectric and piezoelectric effect. The soft materials neither have to be piezoelectric nor magnetostrictive. The outline of this chapter is as follows. In the next section, we briefly summarize the central concept while in Section 3.3, we present the theory and formulation deriving all the necessary equations that fully describe the physical concept. In Section 3.4, we present the numerical solution of the governing equations and discuss the results. Finally, we conclude our work in Section V.

3.2 Central Idea

The central physical concept is depicted in Fig.3.2.1. Consider first a soft homogenous dielectric film as in Fig.3.2.1(a). Neither intrinsic magnetoelectricity nor piezoelectricity exists since the material considered is homogeneous, non-piezoelectric and non-magnetostrictive. Initially, the film is in an undeformed state. In Fig.3.2.1(b), the film is sandwiched between two electrodes and exposed to an external electric field. The film will become polarized

and because of the electric Maxwell's stress, it will tend to compress. If now, an external magnetic field is imposed on this structure (where a pre-existing electric field exists), the magnetic Maxwell's stress will deform the dielectric further, thus creating a change in the pre-existing electric field. The change in the pre-existing electric field due to the presence of the magnetic field manifests itself as the magnetoelectric effect. This mechanism was recently proposed by Liu and Sharma [6]. Several interesting details are notable: (1) Engineering the magnetoelectric effect, based on the nonlinear Maxwell stress concept, is universal in the sense that it occurs in all dielectric films and as a result, any dielectric material can be used. (2) For this mechanism to work, the material must be mechanically soft. This is understandable since the Maxwell stress is significant only for soft polymers. (3) There must exist a magnetic permeability mismatch between the dielectric film and ambient medium. This can be readily ensured by incorporating very minute amounts (negligible from a processing viewpoint) of highly permeable particles (e.g., soft magnetites). (4) Finally, the presence of a pre-existing electric field is required.

In Fig.3.2.1(c), we consider two layers of non-piezoelectric polymers. An electret material is created by incorporating charges and/or dipoles. Among others, this can be accomplished in the following two ways: either by using layers of different materials with charges at the interfaces as in the figure, or by inserting voids in a uniform polymer matrix, i.e. using a foamy material containing charges on void surfaces [9]. Such electret states have been found to be (surprisingly) stable, specifically in certain kinds of polymer foams. The electret material behaves like an apparent piezoelectric material as long as the elastic properties in the material are non-uniform. The mechanism, reminiscent of the magnetoelectric effect discussed in the preceding paragraphs is also based on the nonlinear Maxwell stress effect, deformation of the sample, induces a change in its pre-existing macroscopic polarization manifesting as an apparent piezoelectric effect. Indeed, experimentally, a two-way linear coupling between stress-electric fields has been shown for such electret materials even though microscopically the cause is the Maxwell stress effect rather than any intrinsic piezoelectric property [58, 59]. Polypropylene foams, with fairly stable electret configurations, have been fabricated that exhibit an apparent piezoelectric coefficient six times that of ceramic PZT [58, 59].

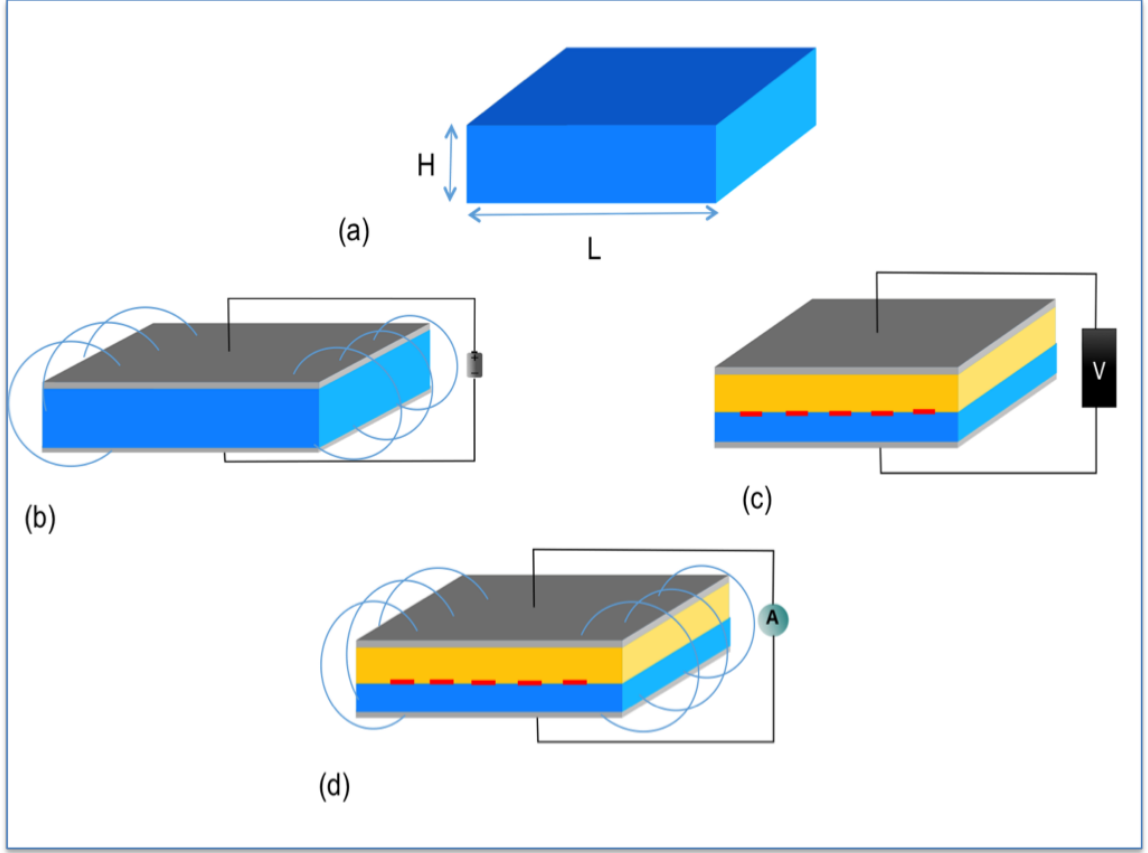


Figure 3.2.1: The central physical concept.

In Fig.3.2.1(d), we combine the previous two cases. This is the central theme of the current chapter. Instead of a pre-existing applied voltage on a homogenous material subject to a magnetic field, we consider a heterogeneous electret material subject to an external magnetic field. The presence of deposited charges inside the soft material will act as an alternative way of applying an electric field, albeit a non-uniform one. The magnetic Maxwell's stress will tend to compress the two layers. This in turn will alter the electric field due to the presence of static charges. As a result, a magnetoelectric effect will be created. In this scheme, we note that an external voltage need not be present for the ME effect to occur. Evidently, as discussed in the case of Fig.3.2.1(c), a piezoelectric will also be present. In other words, Fig.3.2.1(d) represents, schematically, a simple way to create simultaneous magnetoelectric and piezoelectric effect in soft materials. In the following, we present a quantitative explanation of our central idea and a nonlinear theory that can be utilized to

design such materials.

3.3 Theory and Formulation

To derive the governing equations, we consider the simplest possible configuration of an electret material: two layers of soft materials with charges embedded at the interface. The materials forming the two layers are assumed to be elastically non-linear, but electrostatically and magnetostatically linear. We will assume in this context that each constituent material forming the electret structure is isotropic, a valid assumption for soft polymers. The constitutive relations of each of the films are given by

$$\tilde{\Sigma}_{mech} = \frac{\partial W_{elst}(\mathbf{F})}{\partial \mathbf{F}} \quad ; \quad \mathbf{e} = \frac{\mathbf{p}}{\epsilon_0(\epsilon_r - 1)} \quad ; \quad \mathbf{h} = \frac{\mathbf{m}}{\mu_r - 1}, \quad (3.3.1)$$

where $\tilde{\Sigma}_{mech}$, $\mathbf{e}$ and $\mathbf{h}$ are respectively the mechanical Piola-Kirchhoff stress, electric field and magnetic field; $\mathbf{F}$, $\mathbf{p}$ and $\mathbf{m}$ denote the deformation gradient, polarization and magnetization respectively. $W_{elst}(\mathbf{F})$ stands for the elastic energy density, and μ_r, ϵ_r correspond to relative magnetic permeability and electric permittivity respectively.

For the sake of simplicity, we assume that the electrodes are mechanically trivial, and they maintain a constant voltage difference across the films. Assuming the layers to be thin, the electric field in the ambient medium can be safely neglected. Moreover, the magnetic field is considered uniform in the films. Two different sets of coordinates will be used for each layer to maintain the distinction between the reference and the current configurations: $\{X, Y, Z\}$ are the Lagrange coordinates of the material points and $\{x, y, z\}$ are the Euler coordinates.

In this section, we proceed as follows: First, we present the case of a single soft material layer subject to electric and magnetic fields, as explained above, then we extend our results to the elastically heterogeneous electret structure taking into consideration the appropriate boundary conditions.

We consider the following class of deformations $\chi = \chi(X, Y, Z)$:

$$x = X + u(X) \quad ; \quad y = Y(1 + \epsilon_y(X)) \quad ; \quad z = Z(1 + \epsilon_z(X)), \quad (3.3.2)$$

$u, \epsilon_y, \epsilon_z : (0, L) \rightarrow \mathcal{R}$ are scalar functions describing the deformed state of the film. The stretches in the X, Y, Z directions respectively are given by:

$$\lambda_x = 1 + \frac{\partial u}{\partial X} \quad ; \quad \lambda_y = \frac{\partial y}{\partial Y} = 1 + \epsilon_y \quad ; \quad \lambda_z = \frac{\partial z}{\partial Z} = 1 + \epsilon_z. \quad (3.3.3)$$

We denote the deformation gradient by $\mathbf{F} = \text{Grad}\chi$ and the Jacobian measuring the change in the volume by J. In the current configuration, the polarization $\mathbf{p}$ and the magnetization $\mathbf{m}$ are related to the ones defined in the reference configuration by [6, 51]:

$$\mathbf{p} = \frac{\widetilde{\mathbf{P}}}{J} \quad , \quad \mathbf{m} = \frac{\widetilde{\mathbf{M}}}{J}. \quad (3.3.4)$$

We assume that $x(0) = 0$ and $x(H) = h$. The electric field and magnetic field in the current configuration must satisfy the Maxwell's equations with the following boundary conditions:

$$\begin{aligned} \text{div}(-\epsilon_0 \text{grad}\xi + \mathbf{p}) &= 0 \\ \text{div}(-\text{grad}\zeta + \mathbf{m}) &= 0 \\ \xi(x=l) &= V, \quad \xi(x=0) = 0, \\ -\text{grad}\zeta &\longrightarrow \mathbf{h}^e \quad \text{for } x \rightarrow \infty, \end{aligned} \quad (3.3.5)$$

where ξ is the electric potential field, ζ is the magnetic potential field and h^e is the external applied magnetic field.

The stored energy density function of the material is [51, 60]

$$\psi(\mathbf{F}, \widetilde{\mathbf{P}}, \widetilde{\mathbf{M}}) = W_{\text{elst}}(\mathbf{F}) + \frac{|\widetilde{\mathbf{P}}|^2}{2\epsilon_0(\epsilon_r - 1)J} + \frac{\mu_0|\widetilde{\mathbf{M}}|^2}{2(\mu_r - 1)J}. \quad (3.3.6)$$

The first term on the right hand side refers to the elastic energy density whereas the second and third terms describe the contributions of the existence of polarization and magnetization

to the total stored potential energy density. The central idea does not depend on the choice of the elasticity model. For illustration, we employ the Neo-Hookean hyperelasticity model, which models polymers with very limited compressibility [61]:

$$W_{\text{elst}} = \frac{\mu}{2} \left[J^{-\frac{2}{3}} (\lambda_x^2 + \lambda_y^2 + \lambda_z^2) - 3 \right] + \frac{\kappa}{2} (J - 1)^2, \quad (3.3.7)$$

where μ , κ stand respectively for the shear and bulk moduli.

Taking the applied voltage and magnetic field into consideration, the total free energy of the system can be written as [51, 62]:

$$\begin{aligned} \frac{1}{L^2} F[\chi, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}] &= \int_0^H \psi(\mathbf{F}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}) dX \\ &+ \frac{\epsilon_0}{2} \int_0^h \lambda_y \lambda_z |\text{grad} \xi|^2 dx + V \lambda_y \lambda_z (-\epsilon_0 \text{grad} \xi + \mathbf{p})|_{x=0}^{x=h} \\ &+ \int_0^h \lambda_y \lambda_z \frac{\mu_0}{2} |\text{grad} \zeta^{\text{self}}|^2 dx - \int_0^h \lambda_y \lambda_z \mu_0 \mathbf{h}^e \mathbf{m} dx, \end{aligned} \quad (3.3.8)$$

where $-\text{grad} \zeta^{\text{self}} = -\text{grad} \zeta - \mathbf{h}^e$ is the self magnetic field induced by the film alone. The equilibrium state of the film follows from the principle of minimum free energy. Therefore the following variational problem is used [63, 64]:

$$\min_{(\chi, \tilde{\mathbf{P}}, \tilde{\mathbf{M}})} F[\chi, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}]. \quad (3.3.9)$$

Carrying out the appropriate variational calculations with respect to deformation, polarization and magnetization $(\chi, \tilde{\mathbf{P}}, \tilde{\mathbf{M}})$ yields three equilibrium Euler-Lagrange equations. In particular, we find the equilibrium polarization and magnetization satisfy the following relations in the current configuration:

$$\begin{aligned} \mathbf{p} &= -\epsilon_0(\epsilon_r - 1) \text{grad} \xi, \\ \mathbf{m} &= (\mu_r - 1)(-\text{grad} \zeta^{\text{self}} + \mathbf{h}^e), \end{aligned} \quad (3.3.10)$$

which are consistent with eq. (3.3.1) as expected. Inserting these two relations back in the Maxwell's equations (3.3.5), we find the following expressions for the electric field and

polarization:

$$-\text{grad}\xi = -\frac{V}{h}\mathbf{e}_x \quad \text{and} \quad \mathbf{p} = -\epsilon_0(\epsilon_r - 1)\frac{V}{h}\mathbf{e}_x. \quad (3.3.11)$$

Also, if an external magnetic field is applied along the x direction, the self magnetic field and magnetization for $x \in (0, h)$ are given by:

$$-\text{grad}\zeta^{\text{self}} = -\frac{\mu_0 - 1}{\mu_r}h_x^e\mathbf{e}_x \quad ; \quad \mathbf{m} = \frac{\mu_0 - 1}{\mu_r}h_x^e\mathbf{e}_x, \quad (3.3.12)$$

whereas if the magnetic field is applied along the y direction, we obtain

$$-\text{grad}\zeta^{\text{self}} = 0 \quad ; \quad \mathbf{m} = (\mu_r - 1)h_y^e\mathbf{e}_y. \quad (3.3.13)$$

After some algebra, we can rewrite the free energy in terms of $\lambda_x = h/H$ and J as

$$\frac{F}{\mu HL^2} = \hat{W}_{\text{elst}}(\lambda_x, J) - \frac{J}{2\lambda_x^2}\hat{f}^{\text{elect}} - \frac{J}{2}\hat{f}^{\text{mgf}} \quad (3.3.14)$$

with the following abbreviations used for convenience:

$$\begin{aligned} \hat{f}^{\text{elect}} &= \frac{\epsilon_0\epsilon_r V^2}{\mu H^2} \quad , \quad \hat{W}_{\text{elst}} = \frac{1}{\mu}W_{\text{elst}} \\ \hat{f}^{\text{mgf}} &= \frac{\mu_0}{\mu} \left[\frac{\mu_r - 1}{\mu_r} (h_x^e)^2 + (\mu_r - 1)(h_y^e)^2 \right]. \end{aligned} \quad (3.3.15)$$

Now, we extend the model to account for the electret material taking into consideration the appropriate boundary conditions. The equilibrium conditions are then found by minimizing the total free energy $F_T(\lambda_1, \lambda_2, J_1, J_2)$ of the system with respect to variables λ_1, λ_2 that stand respectively for the stretches in the x direction for the first and second layers, and J_1, J_2 that refer to the volume changes of layers 1 and 2. The total free energy is given as

follows:

$$\begin{aligned}
F_T(\lambda_1, \lambda_2, J_1, J_2) = & \int_0^{H_1} \psi_1(\mathbf{F}_1, \widetilde{\mathbf{P}}_1, \widetilde{\mathbf{M}}_1) dX_1 + \int_0^{H_2} \psi_2(\mathbf{F}_2, \widetilde{\mathbf{P}}_2, \widetilde{\mathbf{M}}_2) dX_2 \\
& + \frac{\epsilon_0}{2} \int_0^{h_1} \frac{J_1}{\lambda_1} |\text{grad} \xi_1|^2 dx_1 + V \frac{J_1}{\lambda_1} (-\epsilon_0 \text{grad} \xi_1 + \mathbf{p}_1)|_{X_1=0}^{x_1=h_1} \\
& + \frac{\epsilon_0}{2} \int_0^{h_2} \frac{J_2}{\lambda_2} |\text{grad} \xi_2|^2 dx_2 + V \frac{J_2}{\lambda_2} (-\epsilon_0 \text{grad} \xi_2 + \mathbf{p}_2)|_{X_2=0}^{x_2=h_2} \quad (3.3.16) \\
& + \int_0^{h_1} \frac{J_1}{\lambda_1} \frac{\mu_0}{2} |\text{grad} \zeta_1^{\text{self}}|^2 dx_1 - \int_0^{h_1} \frac{J_1}{\lambda_1} \mu_0 \mathbf{h}^e \cdot \mathbf{m}_1 dx_1 \\
& + \int_0^{h_2} \frac{J_2}{\lambda_2} \frac{\mu_0}{2} |\text{grad} \zeta_2^{\text{self}}|^2 dx_2 - \int_0^{h_2} \frac{J_2}{\lambda_2} \mu_0 \mathbf{h}^e \cdot \mathbf{m}_2 dx_2.
\end{aligned}$$

The boundary conditions emerge from the field equations imposed by Maxwell's equations and the presence of the charges at the interface between the two materials as follows:

$$\begin{aligned}
x(-H_2) = -h_2 \quad ; \quad x(H_1) = h_1 \\
\xi(X=0) = 0 \quad ; \quad \xi(X=H_1) = \xi_1 \quad ; \quad \xi(X=-H_2) = -\xi_2 \quad (3.3.17) \\
\text{div}(-\epsilon_0 \text{grad} \xi + \mathbf{p}) = \rho(x).
\end{aligned}$$

The electric field along the $\mathbf{e}_x$ direction in each of the layers is given by

$$E_1 = -\frac{\xi_1}{h_1} \quad ; \quad E_2 = \frac{\xi_2}{h_2}. \quad (3.3.18)$$

Consequently, we obtain

$$\begin{aligned}
\xi_1 - \xi_2 = V \quad \text{and} \\
D_1 - D_2 = -\frac{\epsilon_1 \epsilon_0 J_1}{\lambda_1^2} \frac{\xi_1}{H_1} - \frac{\epsilon_2 \epsilon_0 J_2}{\lambda_2^2} \frac{\xi_2}{H_2} = q_0, \quad (3.3.19)
\end{aligned}$$

where D_1, D_2 are the electric displacement along the e_x direction.

Using the above interface conditions for the electric field, we find expressions for ξ_1, ξ_2 which are later replaced in the governing equations

$$\begin{aligned}
\frac{\xi_1}{H_1} &= \frac{\epsilon_2 J_2 \lambda_1^2 V - \lambda_1^2 \lambda_2^2 H_2 q_0}{\epsilon_1 J_1 \lambda_2^2 H_2 + \epsilon_2 J_2 \lambda_1^2 H_1} \\
\frac{\xi_2}{H_2} &= \frac{-\epsilon_1 J_1 \lambda_2^2 V - \lambda_1^2 \lambda_2^2 H_1 q_0}{\epsilon_1 J_1 \lambda_2^2 H_2 + \epsilon_2 J_2 \lambda_1^2 H_1}. \quad (3.3.20)
\end{aligned}$$

As for the magnetic field boundary conditions, the following apply:

$$\begin{aligned}\mu_{r_1}(-grad\zeta_1)_x &= \mu_{r_2}(-grad\zeta_2)_x \\ (-grad\zeta_1)_y &= (-grad\zeta_1)_y.\end{aligned}\tag{3.3.21}$$

Minimizing (3.3.16), taking the above equations (3.3.17)-(3.3.21) into consideration, we obtain the following governing equations:

$$\begin{aligned}J_i^{-\frac{2}{3}}\lambda_i^4 - J_i^{\frac{1}{3}}\lambda_i + J_i\hat{f}_i^{elect} &= 0 \\ -\frac{1}{3}(J_i^{-\frac{5}{3}}\lambda_i^4 - J_i^{-\frac{2}{3}}\lambda_i) + \frac{\kappa_i}{\mu_i}(J_i - 1)\lambda_i^2 - \frac{1}{2}\hat{f}_i^{elect} - \frac{1}{2}\hat{f}_i^{mgf}\lambda_i^2 &= 0,\end{aligned}\tag{3.3.22}$$

where

$$\begin{aligned}\hat{f}_i^{elect} &= \frac{\epsilon_i\epsilon_0}{\mu_i}\left(\frac{\xi_i}{H_i}\right)^2 \\ \hat{f}_i^{mgf} &= \frac{\mu_0}{\mu_i}\left[\frac{\mu_{r_i} - 1}{\mu_{r_i}}(h_x^e)^2 + (\mu_{r_i} - 1)(h_y^e)^2\right]\end{aligned}\tag{3.3.23}$$

i is an index which takes values of 1 and 2, used to denote each layer. Finally, the equilibrium state for the system can be found by solving the four equations in (3.3.22).

3.4 Results and discussion

We present the full numerical solution for the four equations in (3.3.22). The materials chosen to illustrate the results are microporous polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF). The materials properties used in the calculations are listed below and can be found from the literature [65, 66, 67]:

Layer 1: $\mu_1 = 0.1MPa$, $\epsilon_1 = 1.5\epsilon_0$, $\kappa_1 = \frac{14}{3}\mu_1$ (corresponding to a Poisson ratio 0.4)

Layer 2: $\mu_2 = 2GPa$, $\epsilon_2 = 9.5\epsilon_0$, $\kappa_2 = \frac{14}{3}\mu_2$ (corresponding to a Poisson ratio 0.4)

The magnetic permeability μ_r of both films is assumed to be 5. The typical value of this parameter, for most polymers, is close to unity. However, as stated earlier, even a minute amount of magnetite particles (which will have negligible mechanical consequences) can easily raise this value to 5 or larger. The charge distribution at the interface is taken to 1 mC/m², a typical value that can be realized experimentally.

Comparing the shear modulus for the two layers, it is found that the microporous PTFE

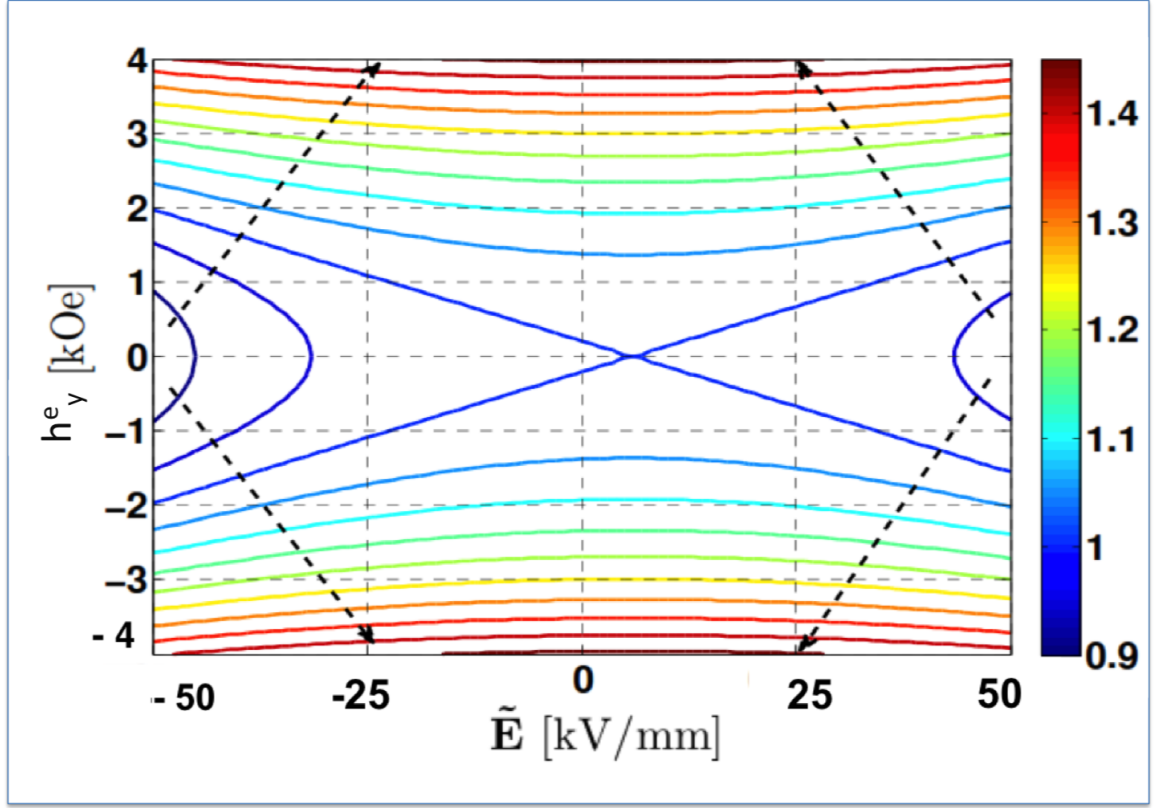


Figure 3.4.1: Contour plot showing the deformation of the dielectric film as a function of the applied electric and magnetic fields.

layer is much softer than the PVDF layer. Thus for the presented model, if the electret is deformed, the deformation will exist mostly within the soft layer. Equations (3.3.22) obviously indicate that the applied electric field $\tilde{E}$ which is along the x-direction, and magnetic field $\mathbf{h}^e = h_y^e \mathbf{e}_y$ is coupled with the deformation. In Fig.3.4.1, we present the strain contour plot with respect to the applied electric and magnetic fields. Here we only show the strain λ_1 of the soft layer with respect to $\tilde{E}$ and h_y^e . The deformation is measured as the ratio of the deformed film thickness subsequent to the application of an external magnetic or electric field and the initial thickness of the film. The arrows indicate the direction in which the deformation increases. From the figure, it is interesting to observe that the effects of $\tilde{E}$ and h_y^e on the deformation are in an opposite sense to each other. The effect of $\tilde{E}$ is to compress the polymer film while h_y^e tends to stretch the film. It is worthwhile to mention

that the contour plot is asymmetric about the line $\tilde{E} = 0$ because of the inserted charges at the interface. Calculations were performed for electric fields around 10^7 V/m, which is below the dielectric strength of both PVDF and PFTE (around 10^8 V/m).

Since the electric and the magnetic fields couple to each other through the deformation, it is also instructive to investigate the effective piezoelectric coefficient of the electret material in the presence of an external magnetic field. Recently, the piezoelectricity of electret structure was investigated [9]. It was shown that for electric fields as high as 0.05 V/nm, the effective piezoelectric response obtained is around twice as that of the Barium Titanate (78 pC/N). In this context, we proceed with the calculation of the piezoelectric coefficient. This can be done by finding the difference between the change in the total thickness as function of the change in the applied voltage for the two cases $q_0 = 0$ and $q_0 \neq 0$. Fig.?? shows the results for various magnetic fields applied. The apparent piezoelectricity of the electret structure is normalized by that of Barium Titanate. Because of the large deformation and the ensuring nonlinearity, the piezoelectric coefficient depends on the applied field, changes nonlinearly with respect to $\tilde{E}$. With the increase of $\tilde{E}$, the piezoelectric coefficient also increases. Recall that the effects of $\tilde{E}$ and h_y^e are in opposite direction, so, as is observed in Fig.3.4.2, not only the piezoelectric response changes, but also its rate of change with respect to $\tilde{E}$ reduces with the existence of magnetic field.

In Fig.3.4.3, we show that the magnetoelectric coupling of the electret is also affected by the inserted charges. In this case, we set the applied electric field $\tilde{E}$ to 10^7 V/m. The actual electric field E is calculated based on the deformation caused by the applied fields $\tilde{E}$ and h_y^e . The magnetoelectric coupling constant dE/dh^e is affected by the interface charges. However, this effect is not significant when the charge density is below 1 mC/m^3 .

The presence of the interface charges can also be thought of as an alternative way of applying an electric field to the electret structure. The inserted charges generate their electric field within the material. Thus, when a magnetic field is applied, the deformation imposed by the magnetic Maxwell stress alters the internal electric field. As a result, a magnetoelectric response is found. Notably, creating an electret structure allows the presence of a magnetoelectric behavior even without the presence of an external voltage source.

In Fig.3.4.4, we study the impact of the volume fraction of the soft layer on the magne-

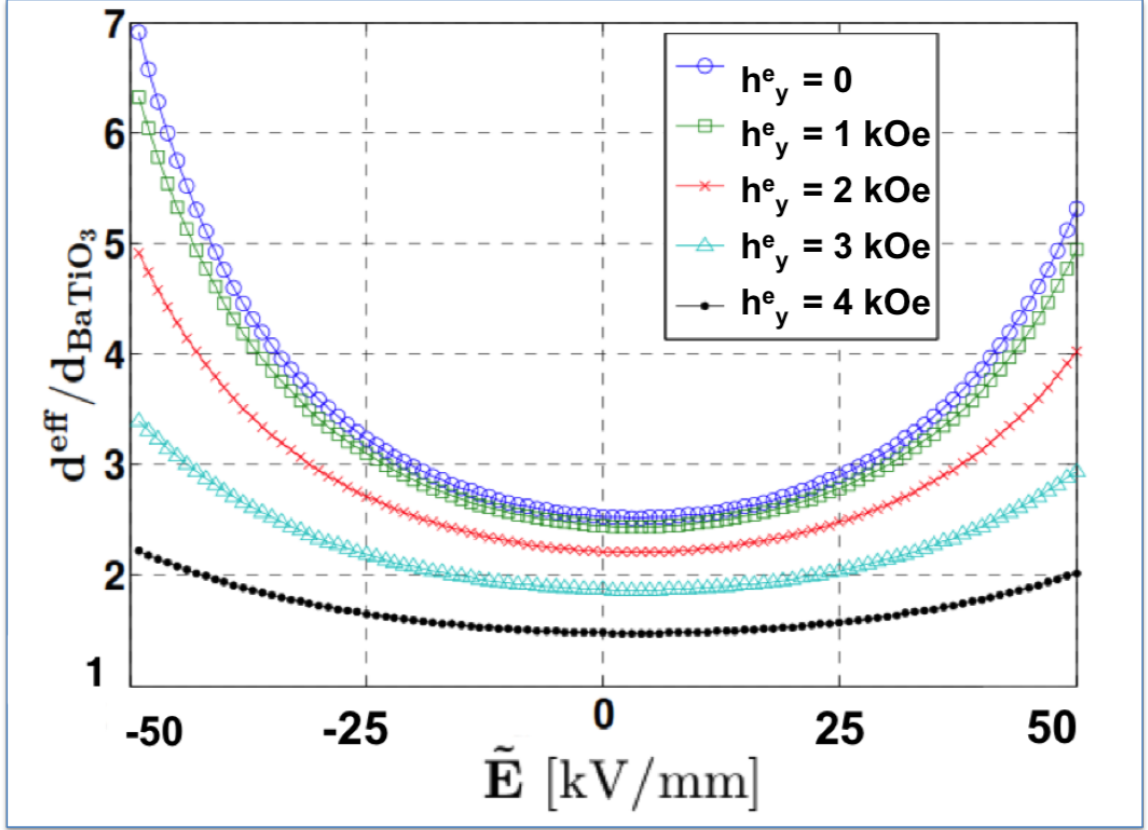


Figure 3.4.2: Effective piezoelectricity of a soft electret material.

toelectric coupling behaviors of an electret. Fig.3.4.4 shows that as the volume fraction of the soft layer increases, the overall magnetoelectric coupling constant dE/dh^e increases correspondingly. Evidently, the large deformation of the soft layer contributes significantly to the magnetoelectric coupling. It is important to mention that this large deformation only exists in very soft materials such as the microporous PTFE used here. From this figure, we also find that for the cases with $\phi \geq 0.5$, dE/dh^e changes nonlinearly with respect to the applied magnetic field h_y^e .

Several implications may be drawn from our results. As seen in Fig.3.4.1, we note that deformations as high as 40% can be achieved for high electric and magnetic fields. While not quite as large as elastomers, the contrast to ceramic-based analogous multifunctional materials is striking¹. How soft should the material be? The upper bound on Maxwell

¹The use of elastomers, unfortunately, does not appear to be practical since charges are unlikely to remain stable for too long in structures made from those.

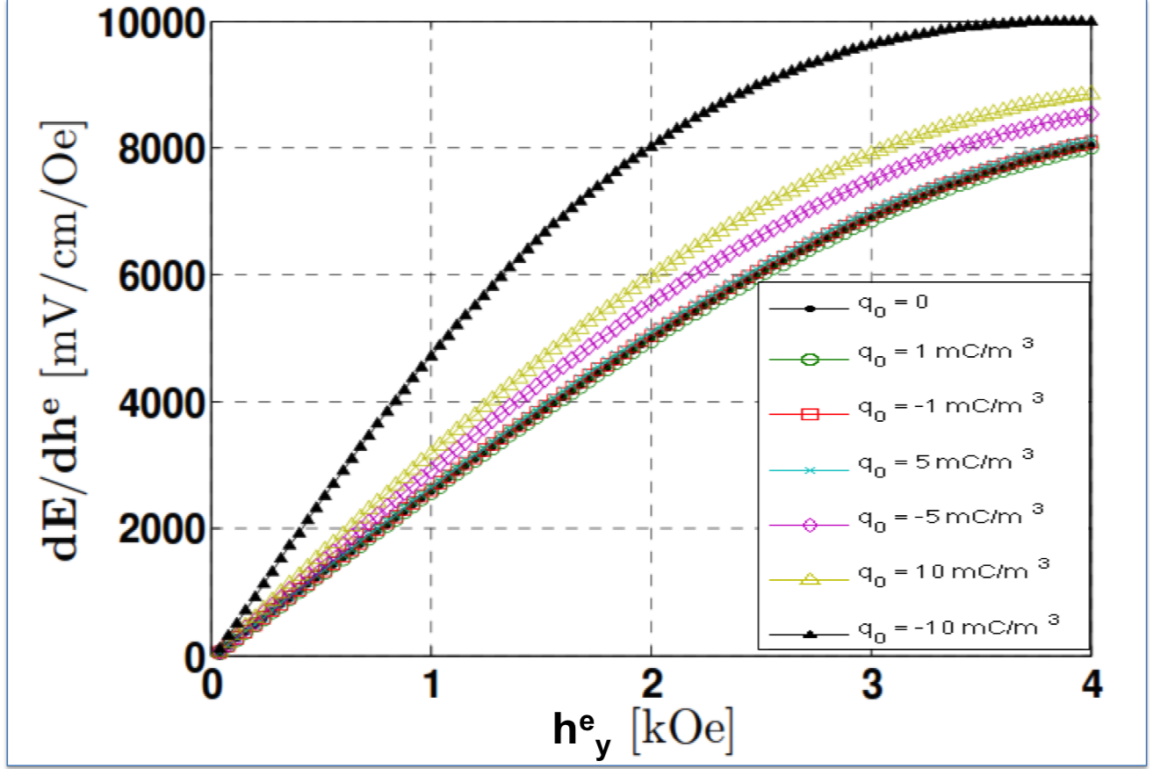


Figure 3.4.3: ME response of the soft electret material with different interface charges.

stress can reach roughly 10^5 Pa if we assume a typical magnetic field on the order of 10^4 Oe and electric field in the range of 10^6 - 10^8 V/m. This translates to about 10% strain induced in a dielectric with a Young modulus of around 1 MPa.

Moreover, this electret structure reveals a large apparent piezoelectric coefficient that depends non-linearly on both the applied electric and magnetic fields. In other words, piezoelectric response can be modified with the application of an external field. The well-known and widely used piezoelectric ceramic material PZT has a linear piezoelectric constant around 290 pC/N which does not depend on any external conditions. Fig.3.4.2 shows that values of this magnitude can be realized for intrinsically non-piezoelectric soft polymer structure by applying appropriate external fields. This comes with the advantage of large deformation that ceramic materials are incapable of. Taking into consideration the results of Fig.3.4.4 that shows the dependence of the magnetoelectric response on the volume fraction of the softer layer, the problem of choosing the appropriate materials for a device

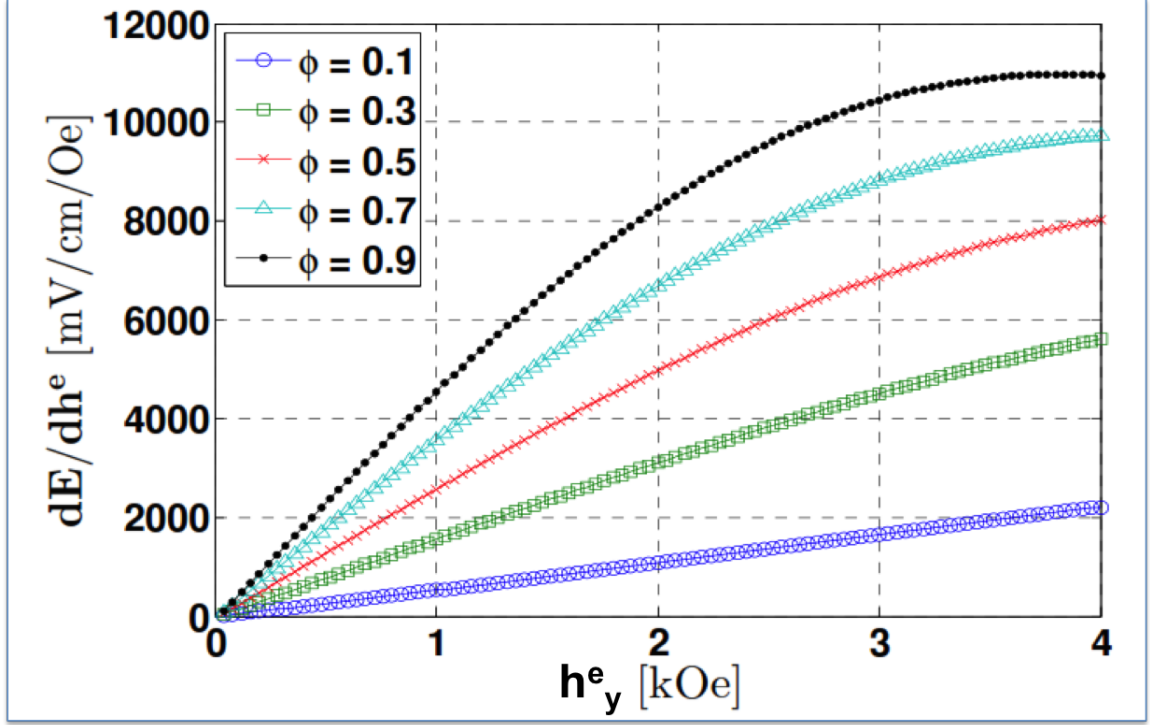


Figure 3.4.4: ME response of the soft electret material with different ϕ

fabrication can be thought of as an optimization problem; picking the polymers that make up the electret structure and the corresponding volume fractions that yield the required piezoelectric or magnetoelectric behavior.

3.5 Conclusion

We propose a relatively simple concept to design soft materials that simultaneously exhibit some rather contradictory properties not usually found in soft materials—apparent piezoelectricity and magnetoelectricity. Geometric nonlinearity and the presence of stable charges within the interfaces of the material is the key mechanism that couples mechanical, electric and magnetic fields. Our work is conceptual and we have illustrated our work using materials that have been experimentally shown to exhibit stable electret structures. Very large deformation may be possible by combining the concepts outline herein with the use of elastomers. However, the latter are likely to be quite vulnerable to charge leakage

—accordingly, experimental work that can find avenues to stabilize electret structures in elastomers is a promising research direction. Extension of our work to complex configurations and three dimensional deformation states appears to be a tantalizing future direction that may pave the way for engineering and optimizing multifunctional material properties through electrets.

Chapter 4

Solid and Liquid Inclusions in Soft Materials: Capillary Effects and Enhanced Electromagnetomechanical Response

4.1 Introduction

Soft materials are currently attracting a growing attention and are intensively researched especially those capable of large mechanical deformations under the application of an external stimulus (such as electric field, magnetic field, temperature,...). For instance, soft materials and more precisely dielectric elastomers that demonstrate strong electromechanical coupling, are considered very promising to host many exciting technological applications such as robotics, actuators, energy harvesting, and stretchable electronics [68, 69, 70, 71, 72, 73, 74, 38, 35, 12, 40, 75, 76, 77].

However, these low permittivity soft dielectrics needs a high electric field to be actuated

(>100 MV/m) [68], and they won't respond to a magnetic field stimulus either. One way to go around this is to create composites by adding micro to nano-sized high permittivity and/or magnetic inclusions into the polymeric matrix. Previous work [78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88] showed that the addition of high permittivity inclusions has proven to enhance the electrostriction of the overall structure. Similarly, if the fillers are magnetic inclusions, then magnetostriction will be observed upon application of an external magnetic field that will act on each particle leading to microscopic deformation of the material and consequently to changing of its shape. As a result of these designs, the deformation of these composites can be controlled by the application of either an electric field or a magnetic field.

One may raise the following question, what if the fillers are magnetic and have also a high permittivity? Well, then we will observe a very interesting phenomenon called magnetoelectricity. But before going further, let us focus on defining magnetoelectricity and why it is so appealing. Single phase magnetoelectric materials has the ability to electrically polarize under the application of a magnetic field, and conversely, magnetize under the application of an electric field. Several applications are investigated based on this tantalizing ability such as wireless energy transfer [46], spintronics, multiple-state memory bits [43], and nonvolatile memories among others [89]. Unfortunately, these materials are rare and restricted to a small set of hard crystalline alloys which limit their use in the proposed applications. However, recent work has shown that this limitation can be circumvented by revealing that soft, highly deformable magnetoelectric materials may be created using a strain-mediated mechanism [6, 49, 48]. The aforementioned mechanism is true for all dielectric materials, does not require the materials to be magnetostrictive or piezoelectric and instead relies on the universal Maxwell stress effect [90]. This effect is significant for soft material as the mechanical strain due to the maxwell stress is roughly $\sim \frac{1}{2E}(\epsilon|\mathbf{e}_0|^2 + \mu|\mathbf{h}_0|^2)$ where ϵ and μ are respectively the permittivity and the permeability of the material, $\mathbf{e}_0$ and $\mathbf{h}_0$ indicate the electric and magnetic fields and E represents the Young's modulus of the dielectric [62, 51, 91]. Based on the latter, we are designing a composite made from a soft dielectric elastomer filled with magnetic, high permittivity inclusions. This composite will not only deform upon application of an electric field and/or a magnetic field but will

also have the amazing ability of generating an electric field upon the action of a magnetic field.

The last but not the least contribution in this chapter is including the surface elasticity. The main purpose of this, is to capture the capillary effects on materials properties at nano and micro-scale. In fact, in order to get the effective properties, many researchers have been relying on the conventional micromechanics to provide a fundamental result describing the composite behavior [29]. However, this doesn't take into account the surface effects at the matrix/inclusion interface and fail to describe the results shown by experiments in case of small scale liquid inclusions embedded into a soft solid. As counterintuitive as this may sounds, these experiments provided the interesting conclusion that adding very soft inclusions to a soft solid will make the overall composite harder [26, 32]. As shown in previous studies [26, 32] and later in this chapter as well, surface effects will alter the elastic properties and will take care of adjusting the elastic solutions to fit this result. In the previous years, the Gurtin-Murdoch model of surface elasticity proposed in 1975 [92] has been extensively used to introduce size effects at the interface inhomogeneity [93, 94]. However, when it comes to soft materials, this model may show some limitations as some material properties used to describe it become negligible and hence compromise the results. Instead, we will start with a fairly simple non linear model, called the capillary surface elasticity model, in which we do not postulate anything beyond surface tension [95]. A careful linearization around the reference state, will give rise to a surface elasticity model that depends only on surface tension which has been previously documented and is not negligible for soft materials. Moreover, While the main interest before was to come up with an enhanced size dependent elastic properties, We see an opportunity here to investigate how capillary effects will impact the electromagnetomechanical response of such composite.

Our aim in this chapter reduces to including surface effects while generating a solution for the magnetoelectric response of a two-phase composite in terms of the electromagnetomechanical properties of the matrix and the inclusions. Magnetostrictive properties will also be deduced in the special case where there is no applied electric field. These closed-form solutions were derived in the limit of small deformations, moderate electric and magnetic fields for the case of an isotropic distribution of spherical inclusions embedded into a matrix.

Moreover, in order to get useful insights into the derived solutions and to come up with possible experiments, specific results were presented and plotted for a dielectric elastomer filled with different elastic, dielectric and magnetic properties including stiff magnetic high permittivity particles and magnetic liquid-like high permittivity inclusions.

4.2 Nonlinear Continuum theory for magneto-electro-elasticity of soft materials

In this section, we will discuss nonlinear continuum theory to explore magnetoelectric coupling behavior for a deformable body. **Notation.** For brevity, wherever possible, we employ direct notation. Vectors and tensors are denoted by bold symbols such as $\mathbf{u}, \mathbf{I}$, 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 $\mathbf{A} \cdot \mathbf{B} := \text{Tr}(\mathbf{A}^T \mathbf{B}) = (\mathbf{A})_{ij} (\mathbf{B})_{ij}$. For a domain D , $\bar{f}_D(\cdot)$ denote the average of the integrand on D . From the viewpoint of matrices, the i^{th} row vector of the gradient of a vector field, e.g., $\nabla \mathbf{u}$, is the gradient of the i^{th} 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, $\text{div} \nabla \mathbf{u} = \Delta \mathbf{u}$ and $\text{div}[(\nabla \mathbf{u})^T] = \nabla(\text{div} \mathbf{u})$. For a scaling parameter $0 < \varepsilon \ll 1$ and a real number n , $O(\varepsilon^n)$ implies the asymptotic behavior $O(\varepsilon^n)/\varepsilon^n \rightarrow C \neq 0$ as $\varepsilon \rightarrow 0$ whereas $o(\varepsilon^n)/\varepsilon^n \rightarrow 0$ as $\varepsilon \rightarrow 0$.

We begin with the kinematics and assume that the thermodynamic state of the body is described by the deformation, nominal polarization and nominal magnetization $(\mathbf{y}, \mathbf{p}, \mathbf{m}) : \mathbf{D} \rightarrow \mathbb{R}^3 \times \mathbb{R}^3$. By the Maxwell equations, we introduce the electric and magnetic potentials ξ and ζ respectively.

For simplicity, we consider the electric Dirichlet boundary conditions

$$\xi = \xi_b \quad \text{on } \partial D \quad \text{and} \quad \mathbf{y} = \mathbf{x} + \mathbf{u}_b \quad \text{on } \partial D, \quad (4.2.1)$$

where $\xi_b : \partial D \rightarrow \mathbb{R}$ (resp. $\mathbf{u}_b : \partial D \rightarrow \mathbb{R}^3$) is the imposed boundary potential (resp. boundary displacement).

The magnetic boundary condition is given by

$$-\nabla\zeta \rightarrow \mathbf{h}^e \quad \text{as} \quad |\mathbf{x}| \rightarrow +\infty. \quad (4.2.2)$$

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 electric Maxwell equation can be written as (ϵ_0 is the vacuum permittivity)

$$\operatorname{div}_{\mathbf{y}}(-\epsilon_0 \nabla_{\mathbf{y}} \xi + \mathbf{p}/J) = 0 \quad \text{in } \mathbf{y}(D), \quad (4.2.3)$$

where subscript $\mathbf{y}$ means that the derivatives are taken with respect to the Eulerian coordinates $\mathbf{y}$.

The magnetic Maxwell's equation is given by

$$\operatorname{div}_{\mathbf{y}}(-\nabla_{\mathbf{y}} \zeta + \mathbf{m}/J) = 0 \quad \text{in } \mathbf{y}(D). \quad (4.2.4)$$

In what follows, to conveniently couple deformation to electrical and magnetic quantities, we work in the *reference* configuration where the Maxwell's equation (4.2.3) is transformed to

$$\nabla \cdot (-\epsilon_0 J \mathbf{C}^{-1} \nabla \xi + \mathbf{F}^{-1} \mathbf{p}) = 0 \quad \text{in } D, \quad (4.2.5)$$

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

The second Maxwell's equation (4.2.4) is transformed to

$$\nabla \cdot (-J\mathbf{C}^{-1}\nabla\zeta + \mathbf{F}^{-1}\mathbf{m}) = 0 \quad \text{in } D. \quad (4.2.6)$$

We also assume that there are no external magnetic moments.

The total free energy of the system is given by [96, 51]

$$F[\mathbf{y}, \mathbf{p}, \mathbf{m}] = \mathbf{U}[\nabla\mathbf{y}, \mathbf{p}, \mathbf{m}] + \mathbf{E}^{elec}[\nabla\mathbf{y}, \mathbf{m}] + \mathbf{E}^{mag}[\nabla\mathbf{y}, \mathbf{m}]. \quad (4.2.7)$$

The first term $\mathbf{U}[\nabla\mathbf{y}, \mathbf{p}, \mathbf{m}]$ refers to the internal energy of the system, the second term $\mathbf{E}^{elec}[\nabla\mathbf{y}, \mathbf{m}]$ refers to the total electric energy and the last term $\mathbf{E}^{mag}[\nabla\mathbf{y}, \mathbf{m}]$ denotes the total magnetic energy.

The internal energy is given by

$$\mathbf{U}[\nabla\mathbf{y}, \mathbf{p}, \mathbf{m}] = \int_D \Psi(\nabla\mathbf{y}, \mathbf{p}) d\mathbf{x}, \quad (4.2.8)$$

where the internal energy density function $\Psi : \mathbb{R}^{3 \times 3} \times \mathbb{R}^3 \rightarrow \mathbb{R}$ prescribes the electro-elastic constitutive laws of the material.

The electric energy is given by

$$\mathbf{E}^{elec}[\nabla\mathbf{y}, \mathbf{m}] = \int_D \frac{\epsilon_0}{2} \nabla\zeta \cdot J\mathbf{C}^{-1}\nabla\zeta d\mathbf{x} + \int_{\partial D} \xi_b \mathbf{n} \cdot (-\epsilon_0 J\mathbf{C}^{-1}\nabla\zeta + \mathbf{F}^{-1}\mathbf{p}) d\mathbf{x}. \quad (4.2.9)$$

We note that the total electric energy consists of the total field energy in addition to the potential energy due to the boundary device, i.e. battery, (the second integral in (4.2.9)) used to maintain the boundary potential. Using the divergence theorem, the two integrals in equation (4.2.9) can be combined into one. The electric energy can be written as

$$\mathbf{E}^{elec}[\nabla\mathbf{y}, \mathbf{m}] = \int_D \left[-\frac{\epsilon_0}{2} \nabla\zeta \cdot J\mathbf{C}^{-1}\nabla\zeta + \nabla\zeta \cdot \mathbf{F}^{-1}\mathbf{p} \right] d\mathbf{x}. \quad (4.2.10)$$

Finally, the total magnetic field energy is given by

$$\mathbf{E}^{mag}[\nabla \mathbf{y}, \mathbf{m}] = \int_D \frac{\mu_0}{2} J |\mathbf{F}^{-T} \nabla \zeta|^2 d\mathbf{x}. \quad (4.2.11)$$

By similar calculations as in [62, 51], we can show that within a constant independent of the state variables $(\mathbf{y}, \mathbf{m}, \mathbf{p})$, the magnetic energy can be written as

$$\mathbf{E}^{mag}[\mathbf{y}, \mathbf{m}] = \int_D \frac{\mu_0}{2} |\mathbf{F}^{-T} \nabla \zeta^{self}|^2 d\mathbf{x} - \int_{\partial D} \mu_0 \mathbf{h}^e \cdot \mathbf{m} d\mathbf{x}, \quad (4.2.12)$$

where $-\zeta^{self} = -\zeta - \mathbf{h}^e$ is the self magnetic field induced by the material and $-\zeta^{self} \rightarrow 0$ as $|x| \rightarrow +\infty$. Thus, the expression for the total energy is given by

$$\begin{aligned} F[\mathbf{y}, \mathbf{p}, \mathbf{m}] &= \int_D \Psi(\nabla \mathbf{y}, \mathbf{p}) d\mathbf{x} \\ &+ \int_D \left[-\frac{\epsilon_0}{2} \nabla \xi \cdot J \mathbf{C}^{-1} \nabla \xi + \nabla \xi \cdot \mathbf{F}^{-1} \mathbf{p} \right] d\mathbf{x} \\ &+ \int_D \frac{\mu_0}{2} |\mathbf{F}^{-T} \nabla \zeta^{self}|^2 d\mathbf{x} - \int_{\partial D} \mu_0 \mathbf{h}^e \cdot \mathbf{m} d\mathbf{x}. \end{aligned} \quad (4.2.13)$$

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

$$\frac{\partial \Psi}{\partial \mathbf{F}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = \frac{\partial \Psi}{\partial \mathbf{p}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = \frac{\partial \Psi}{\partial \mathbf{m}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = 0. \quad (4.2.14)$$

Also, we assume the material does not have any *intrinsic* properties (such as piezoelectricity or magnetoelectricity) in the sense that

$$\frac{\partial^2 \Psi}{\partial \mathbf{F} \partial \mathbf{p}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = \frac{\partial^2 \Psi}{\partial \mathbf{F} \partial \mathbf{m}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = \frac{\partial^2 \Psi}{\partial \mathbf{m} \partial \mathbf{p}} \Big|_{(\mathbf{F}, \mathbf{p}, \mathbf{m})=(\mathbf{I}, 0, 0)} = 0. \quad (4.2.15)$$

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

$$\nabla \mathbf{u} \sim \varepsilon \ll 1, \quad \mathbf{p} \sim \varepsilon^{1/2} \quad (-\nabla \xi \sim \varepsilon^{1/2}), \quad \mathbf{m} \sim \varepsilon^{1/2} \quad (-\nabla \zeta \sim \varepsilon^{1/2}). \quad (4.2.16)$$

Then, as shown by Tian and co-workers (2007, 2012) and Liu (2014), we can formally

decompose the free energy (4.2.13) according to their order of magnitude as compared with the small parameter ε . To this end, by (4.2.16) we recall the algebraic identities

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

Inserting the above equations into (4.2.13), by (4.2.16) we obtain

$$F[\mathbf{y}, \mathbf{p}, \mathbf{m}] = F^{(0)} + F^{(1)} + F^{(2)} + o(\varepsilon^2), \quad (4.2.17)$$

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

$$\begin{aligned}F_p^{(1)}[\mathbf{p}] &= \int_D \left[\frac{1}{2} \mathbf{p} \cdot \boldsymbol{\chi} \mathbf{p} + \frac{\epsilon_0}{2} |\nabla \xi|^2 \right] d\mathbf{x} + \int_{\partial D} \xi_b \mathbf{n} \cdot (-\epsilon_0 \nabla \xi + \mathbf{p}) d\mathbf{x} \sim \varepsilon, \\ F_m^{(1)}[\mathbf{m}] &= \int_D \left[\frac{1}{2} \mathbf{m} \cdot \boldsymbol{\Upsilon} \mathbf{m} + \frac{\mu_0}{2} |\nabla \zeta^{self}|^2 - \mu_0 \mathbf{h}^e \mathbf{m} \right] d\mathbf{x} \sim \varepsilon, \\ F^{(2)}[\mathbf{u}, \mathbf{p}, \mathbf{m}] &= \int_D \left[\frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C} \nabla \mathbf{u} + \nabla \mathbf{u} \cdot \mathbb{M}(\mathbf{p} \otimes \mathbf{p}) + \nabla \mathbf{u} \cdot \mathbb{N}(\mathbf{m} \otimes \mathbf{m}) + \nabla \mathbf{u} \cdot \boldsymbol{\sigma}_{\text{MW}} \right] d\mathbf{x} \sim \varepsilon^2.\end{aligned} \quad (4.2.18)$$

Also, the leading-order Maxwell stress $\boldsymbol{\sigma}_{\text{MW}}$ is given by

$$\boldsymbol{\sigma}_{\text{MW}} = -\frac{\epsilon_0}{2} |\nabla \xi|^2 \mathbf{I} + \epsilon_0 \nabla \xi \otimes \nabla \xi - \nabla \xi \otimes \mathbf{p} - \frac{\mu_0}{2} |\nabla \zeta|^2 \mathbf{I} + \mu_0 \nabla \zeta \otimes \nabla \zeta - \nabla \zeta \otimes \mathbf{m}, \quad (4.2.19)$$

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

$$\boldsymbol{\chi} = \frac{\partial^2 \Psi}{\partial \mathbf{p} \partial \mathbf{p}}, \quad \boldsymbol{\Upsilon} = \frac{\partial^2 \Psi}{\partial \mathbf{m} \partial \mathbf{m}}, \quad \mathbb{C} = \frac{\partial^2 \Psi}{\partial \mathbf{F} \partial \mathbf{F}}, \quad \mathbb{M} = \frac{1}{2} \frac{\partial^3 \Psi}{\partial \mathbf{F} \partial \mathbf{p} \partial \mathbf{p}}, \quad \mathbb{N} = \frac{1}{2} \frac{\partial^3 \Psi}{\partial \mathbf{F} \partial \mathbf{m} \partial \mathbf{m}} \quad (4.2.20)$$

Similarly, we can rewrite equations (4.2.5) and (4.2.6) as

$$\begin{aligned}\nabla \cdot (-\epsilon_0 \nabla \xi + \mathbf{p}) + o(\epsilon^{1/2}) &= 0 \quad \text{in } D, \\ \nabla \cdot (-\nabla \zeta + \mathbf{m}) + o(\epsilon^{1/2}) &= 0.\end{aligned}\tag{4.2.21}$$

Then, according to the principle of minimum free energy, the equilibrium state of the system is determined by the variational principle, together with the constraints (4.2.5) and (4.2.6). This implies the sequential minimization problems

$$\begin{aligned}\min_{\mathbf{p}} \{F_p^{(1)}[\mathbf{p}] : \xi \text{ satisfies (4.2.1)}_1 \text{ and (4.2.21)}_1\}, \text{ and} \\ \min_{\mathbf{m}} \{F_m^{(1)}[\mathbf{m}] : \zeta \text{ satisfies (4.2.2)}_2 \text{ and (4.2.21)}_2\}, \text{ and} \\ \min_{\mathbf{u}} \{F^{(2)}[\mathbf{u}, \mathbf{p}, \mathbf{m}] : \mathbf{u} \text{ satisfies (4.2.1)}_2\}.\end{aligned}\tag{4.2.22}$$

Immediately, we find that the associated Euler-Lagrange equations for $(\mathbf{p}, \mathbf{u}, \mathbf{m})$ are given by

$$\begin{cases} \nabla \xi + \chi \mathbf{p} = 0, \\ \nabla \zeta + \Upsilon \mathbf{m} = 0, \\ \operatorname{div}[\mathbb{C} \nabla \mathbf{u} + \mathbb{M}(\mathbf{p} \otimes \mathbf{p}) + \mathbb{N}(\mathbf{m} \otimes \mathbf{m}) + \boldsymbol{\sigma}_{\text{MW}}] = 0. \end{cases}\tag{4.2.23}$$

Taking into account (4.2.21) to the leading order and eliminating $\mathbf{p}$ and $\mathbf{m}$ by (4.2.23)_{1,2} respectively, we conclude the following system of differential equations for $(\mathbf{u}, \xi, \zeta)$:

$$\begin{cases} \nabla \cdot (\boldsymbol{\epsilon} \nabla \xi) = 0 & \text{in } D, \\ \nabla \cdot (\boldsymbol{\mu} \nabla \zeta) = 0 & \text{in } D, \\ \operatorname{div}[\mathbb{C} \nabla \mathbf{u} + \mathbb{A}(\nabla \xi \otimes \nabla \xi) + \mathbb{B}(\nabla \zeta \otimes \nabla \zeta)] = 0 & \text{in } D, \end{cases}\tag{4.2.24}$$

where $\boldsymbol{\epsilon} = \epsilon_0 \mathbf{I} + \chi^{-1}$ and $\boldsymbol{\mu} = \mu_0(\Upsilon^{-1} + \mathbf{I})$,

$$(\mathbb{A})_{ijkl} = (\mathbb{M})_{ijk'l'}(\chi^{-1})_{kk'}(\chi^{-1})_{ll'} + \frac{\epsilon_0}{2} \mathbb{T}_{ijkl} + \frac{1}{2} [\delta_{ik}(\chi^{-1})_{jl} + \delta_{il}(\chi^{-1})_{jk}], \tag{4.2.25}$$

$$(\mathbb{B})_{ijkl} = (\mathbb{N})_{ijk'l'}(\Upsilon^{-1})_{kk'}(\Upsilon^{-1})_{ll'} + \frac{\mu_0}{2} \mathbb{T}_{ijkl} + \frac{\mu_0}{2} [\delta_{ik}(\Upsilon^{-1})_{jl} + \delta_{il}(\Upsilon^{-1})_{jk}], \tag{4.2.26}$$

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

$$\begin{aligned}\mathbb{T}\mathbf{F} &= \mathbf{F} + \mathbf{F}^T - (\text{Tr}\mathbf{F})\mathbf{I} \quad \forall \mathbf{F} \in \mathbb{R}^{3 \times 3}, \\ \mathbb{T}_{ijkl} &= \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \delta_{ij}\delta_{kl}.\end{aligned}\tag{4.2.27}$$

For an ideal dielectric material whose electric permittivity tensor $\boldsymbol{\epsilon}$ and magnetic permeability tensor $\boldsymbol{\mu}$ are both independent of deformation, the internal energy density function $\Psi = \Psi(\mathbf{F}, \mathbf{p}, \mathbf{m})$ has to be of the following form [51]:

$$\Psi(\mathbf{F}, \mathbf{p}, \mathbf{m}) = W(\mathbf{U}) + \frac{1}{2J}\mathbf{p} \cdot \mathbf{R}\boldsymbol{\chi}\mathbf{R}^T\mathbf{p} + \frac{\mu_0}{2J}\mathbf{m} \cdot \mathbf{R}\boldsymbol{\Upsilon}\mathbf{R}^T\mathbf{m},$$

where $W = W(\mathbf{U})$ is the elastic energy density, $\mathbf{U} = \mathbf{C}^{1/2}$, $\boldsymbol{\chi} = (\boldsymbol{\epsilon} - \epsilon_0\mathbf{I})^{-1}$, $\boldsymbol{\Upsilon} = (\boldsymbol{\mu}/\mu_0 - \mathbf{I})^{-1}$ and $\mathbf{R} = \mathbf{F}\mathbf{U}^{-1} \in SO(3)$. To calculate the tensors $\mathbb{M}$ and $\mathbb{N}$ defined by (4.2.20), we expand the scalar function $(\varepsilon, \eta, \beta) \mapsto \Psi(\mathbf{I} + \varepsilon\mathbf{F}_1, \eta\mathbf{p}_1, \beta\mathbf{m}_1)$ for fixed $(\mathbf{F}_1, \mathbf{p}_1, \mathbf{m}_1) \in \mathbb{R}^{3 \times 3} \times \mathbb{R}^3$ at $(\varepsilon, \eta, \beta) = 0$. Since the material does not assume intrinsic magnetoelectricity, we did the expansions of η and β separately.

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

where we have employed the following algebraic identities:

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

Therefore,

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

and hence the electrostrictive tensor $\mathbb{A}$ defined by (4.2.25) satisfies

$$\mathbb{A}(\nabla\xi \otimes \nabla\xi) = \frac{1}{2}[\nabla\xi \otimes (\epsilon\nabla\xi) + (\epsilon\nabla\xi) \otimes \nabla\xi - \mathbf{I}(\nabla\xi \cdot \epsilon\nabla\xi)]. \quad (4.2.28)$$

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

$$(\mathbb{A})_{ijkl} = \frac{\epsilon}{2}\mathbb{T}_{ijkl}, \quad \text{i.e.,} \quad \mathbb{A}(\nabla\xi \otimes \nabla\xi) = \epsilon\nabla\xi \otimes \nabla\xi - \frac{\epsilon}{2}|\nabla\xi|^2. \quad (4.2.29)$$

Similarly, we do the expansion for β

$$\begin{aligned} \Psi(\mathbf{I} + \varepsilon\mathbf{F}_1, \beta\mathbf{m}_1) &= W(\mathbf{U}_\varepsilon) + \beta^2 \frac{\mu_0}{2} \mathbf{m}_1 \cdot \Upsilon \mathbf{m}_1 \\ &\quad + \frac{\beta^2 \varepsilon}{2} \mu_0 [-\mathbf{m}_1 \cdot \Upsilon \mathbf{m}_1 \text{Tr} \mathbf{F}_1 - \mathbf{F}_1 \cdot (\Upsilon \mathbf{m}_1) \otimes \mathbf{m}_1 + \mathbf{F}_1^T \cdot (\Upsilon \mathbf{m}_1) \otimes \mathbf{m}_1] + o(\beta^2 \varepsilon), \end{aligned}$$

where we have employed the same algebraic identities as above. The last one of them now becomes:

$$\mathbf{m}_1 \cdot \mathbf{R}_\varepsilon \Upsilon \mathbf{R}_\varepsilon^T \mathbf{m}_1 = \frac{\varepsilon}{2} \mathbf{F}_1 \cdot [\mathbf{m}_1 \otimes (\Upsilon \mathbf{m}_1) - (\Upsilon \mathbf{m}_1) \otimes \mathbf{m}_1] + o(\varepsilon).$$

Therefore,

$$\mathbf{F}_1 \cdot \mathbb{N}(\mathbf{m}_1 \otimes \mathbf{m}_1) = \frac{1}{2} \mathbf{F}_1 \cdot [\mathbf{m}_1 \otimes (\Upsilon \mathbf{m}_1) - (\Upsilon \mathbf{m}_1) \otimes \mathbf{m}_1] - \frac{1}{2} \mathbf{m}_1 \cdot \Upsilon \mathbf{m}_1 \text{Tr} \mathbf{F}_1,$$

and hence the magnetostrictive tensor $\mathbb{B}$ defined by (4.2.25) satisfies

$$\mathbb{B}(\nabla\zeta \otimes \nabla\zeta) = \frac{1}{2}[\nabla\zeta \otimes (\mu\nabla\zeta) + (\mu\nabla\zeta) \otimes \nabla\zeta - \mathbf{I}(\nabla\zeta \cdot \mu\nabla\zeta)]. \quad (4.2.30)$$

If the material is isotropic with permeability tensor $\mu = \mu\mathbf{I}$, we have

$$(\mathbb{B})_{ijkl} = \frac{\mu}{2}\mathbb{T}_{ijkl}, \quad \text{i.e.,} \quad \mathbb{B}(\nabla\zeta \otimes \nabla\zeta) = \mu\nabla\zeta \otimes \nabla\zeta - \frac{\mu}{2}|\nabla\zeta|^2. \quad (4.2.31)$$

4.3 Effective Magnetoelectric Properties of Composites With Capillary Effects

We consider a composite model that consists of a single inclusion Ω_0 in a representative volume element $\mathbf{Y} \subset \mathbb{R}$. The inclusion $\Omega_0 \subset \mathbf{Y}$ is assumed to be a sphere of radius R_0 and magnetic permeability $\mu^{\text{in}} > 1$. Physical intuition dictates that soft and highly polarizable materials allow for strong electro-elastic coupling effects. Homogenization and effective electrostrictive properties of this composite in the existence of an external electric field and contribution of surface capillary effects has been discussed in a recent work [78]. The deposition of magnetite particles on the inclusion changes its magnetic permeability, thus making the composite vulnerable to the existence of an external magnetic field as well. In our work, We are interested in investigating the effective properties of the matrix/inclusion composite in the presence of an external electric and magnetic field while accounting for surface capillary effects. In order to explore the effective magneto-electro-elastic properties, We employ the variational formulation and the principle of energy equivalence.

The interface between the matrix and the inclusion $\partial\Omega_0$ is considered as an independent elastic entity which adds an elastic energy term dependent on the configuration. As for the bulk elasticity, we will consider isotropic surfaces and small deformations. Both the matrix and the inclusion are assumed to be linearly elastic, with stiffness tensors $\mathbb{C}^{\text{in}}$ and $\mathbb{C}^{\text{ex}}$ respectively. The reference configuration is defined as the state with zero bulk stress. Since our system of interest is a composite, we need to introduce position dependent materials properties. The x-dependent stiffness tensor $\mathbb{C}_{\#}$ is given by

$$\mathbb{C}_{\#} = \begin{cases} \mathbb{C}^{\text{ex}} & \text{in } Y \setminus \Omega_0 \\ \mathbb{C}^{\text{in}} & \text{in } \Omega_0. \end{cases} \quad (4.3.1)$$

Similarly, the x-dependent electric permittivity $\epsilon_{\#}$ and magnetic permeability $\mu_{\#}$ are given

by

$$\epsilon_{\#} = \begin{cases} \epsilon^{\text{ex}} & \text{in } Y \setminus \Omega_0 \\ \epsilon^{\text{in}} & \text{in } \Omega_0 \end{cases} \quad (4.3.2)$$

and

$$\mu_{\#} = \begin{cases} \mu^{\text{ex}} & \text{in } Y \setminus \Omega_0 \\ \mu^{\text{in}} & \text{in } \Omega_0. \end{cases} \quad (4.3.3)$$

For explicit solutions, we consider a coated sphere and assume that (i) the exterior and interior solids are isotropic with $G^{\text{in}}, \kappa^{\text{in}}$ (resp. $G^{\text{ex}}, \kappa^{\text{ex}}$) being the shear modulus and bulk modulus of the inclusion (resp. matrix), and (ii) the average strain $\bar{\mathbf{E}}$ is prescribed as

$$\bar{\mathbf{E}} = a[\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2] + b\mathbf{e}_3 \otimes \mathbf{e}_3, \quad (4.3.4)$$

where $(\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3)$ are the basis vectors for the canonical cartesian coordinate system.

4.3.1 Surface Energy Contribution

Surface energy contribution is given by

$$\mathbf{E}_s[\mathbf{u}] = \int_{\partial\Omega_0} \frac{1}{2} [\nabla_s \mathbf{u} \cdot \mathbb{C}_s \nabla_s \mathbf{u} - \frac{2\gamma}{R_0} (\mathbf{u} \otimes \mathbf{n}) \cdot \mathbb{T} \nabla_s \mathbf{u}] ds. \quad (4.3.5)$$

In the above expression for capillary energy, ∇_s is the surface gradient, $\mathbb{C}_s$ is the surface stiffness tensor and γ is the surface tension. $\mathbb{T} : \mathbb{R}^{3 \times 3} \rightarrow \mathbb{R}^{3 \times 3}$ is a fourth order tensor given by

$$\begin{aligned} \mathbb{T}\mathbf{F} &= (\text{Tr}\mathbf{F})\mathbf{I} - \mathbf{F} \quad \forall \mathbf{F} \in \mathbb{R}^{3 \times 3}, \\ \mathbb{T}_{ijkl} &= -\delta_{il}\delta_{jk} + \delta_{ij}\delta_{kl}. \end{aligned} \quad (4.3.6)$$

4.3.2 Homogenization formulation and effective magnetostrictive properties

If the average magnetic field in the RVE is prescribed as $\bar{\mathbf{h}} \in \mathbb{R}$, the local magnetic field $-\nabla\zeta_{\bar{\mathbf{h}}} : \mathbf{Y} \rightarrow \mathbb{R}$ is uniquely determined by the variational problem (eq. (4.2.18)):

$$\mathbf{H}^{\text{eff}}(\bar{\mathbf{h}}) := \min\{\mathbf{H}[\zeta] = \int_{\mathbf{Y}} \frac{\mu_{\#}}{2} |\nabla\zeta|^2 : \zeta \in \mathcal{M}_{\bar{\mathbf{h}}}\}, \quad (4.3.7)$$

where $\mathcal{M}_{\bar{\mathbf{h}}}$ is the admissible space for the magnetic potential defined as

$$\mathcal{M}_{\bar{\mathbf{h}}} \equiv \{\zeta : -\oint_Y \nabla\zeta = \bar{\mathbf{h}} \text{ and } \nabla\zeta \text{ is } Y\text{-periodic}\}. \quad (4.3.8)$$

Assuming that $\zeta_{\bar{\mathbf{h}}} \in \mathcal{M}_{\bar{\mathbf{h}}}$ is a minimizer for (4.3.7), then it should satisfy the associated Euler-Lagrange equation

$$\nabla \cdot (\boldsymbol{\mu}_{\#} \nabla \zeta_{\bar{\mathbf{h}}}) = 0 \quad \text{in } \mathbf{Y}. \quad (4.3.9)$$

On the other hand, the effective magnetic permeability tensor of the composite μ^{eff} is determined through the minimum energy $\mathbf{H}^{\text{eff}}(\bar{\mathbf{h}})$ by

$$\mathbf{H}^{\text{eff}}(\bar{\mathbf{h}}) = \frac{1}{2} \bar{\mathbf{h}} \cdot \mu^{\text{eff}} \bar{\mathbf{h}}, \quad \text{i.e.,} \quad \mu^{\text{eff}} = \frac{\partial^2 \mathbf{H}^{\text{eff}}(\bar{\mathbf{h}})}{\partial \bar{\mathbf{h}} \partial \bar{\mathbf{h}}}. \quad (4.3.10)$$

For ideal dielectric materials, the magnetostrictive stress due to the magnetic field defined previously in equations (4.2.31) would be written as

$$\boldsymbol{\sigma}_{\text{mag}} = \mathbb{B}_{\#}(\nabla\zeta_{\bar{\mathbf{h}}} \otimes \nabla\zeta_{\bar{\mathbf{h}}}), \quad (4.3.11)$$

where the fourth order tensor $\mathbb{B}_{\#}$ is given by

$$\mathbb{B}_{\#}(\nabla\zeta \otimes \nabla\zeta) = \mu_{\#} \nabla\zeta \otimes \nabla\zeta - \frac{\mu_{\#}}{2} |\nabla\zeta|^2. \quad (4.3.12)$$

Meanwhile, if we assume $\bar{\mathbf{e}} \in \mathbb{R}$ to be the average electric field in the RVE, then the local electric field $-\nabla \xi_{\bar{\mathbf{e}}} : \mathbf{Y} \rightarrow \mathbb{R}$ is uniquely determined by the variational problem ((4.2.18))

$$\mathbf{G}^{\text{eff}}(\bar{\mathbf{e}}) := \min\{\mathbf{G}[\xi] = \int_{\mathbf{Y}} \frac{\epsilon_{\#}}{2} |\nabla \xi|^2 : \xi \in \mathcal{P}_{\bar{\mathbf{e}}}\}, \quad (4.3.13)$$

where $\mathcal{P}_{\bar{\mathbf{e}}}$ is the admissible space for the electric potential defined as

$$\mathcal{U}_{\bar{\mathbf{e}}} \equiv \{\xi : -\oint_Y \nabla \xi = \bar{\mathbf{e}} \text{ and } \nabla \xi \text{ is } Y\text{-periodic}\}. \quad (4.3.14)$$

Assuming that $\xi_{\bar{\mathbf{e}}} \in \mathcal{P}_{\bar{\mathbf{e}}}$ is a minimizer for (4.3.13), then it should satisfy the associated Euler-Lagrange equation

$$\nabla \cdot (\epsilon_{\#} \nabla \xi_{\bar{\mathbf{e}}}) = 0 \quad \text{in } \mathbf{Y}. \quad (4.3.15)$$

On the other hand, the effective electric permittivity tensor of the composite ϵ^{eff} is determined through the minimum energy $\mathbf{G}^{\text{eff}}(\bar{\mathbf{e}})$ by

$$\mathbf{G}^{\text{eff}}(\bar{\mathbf{e}}) = \frac{1}{2} \bar{\mathbf{e}} \cdot \epsilon^{\text{eff}} \bar{\mathbf{e}}, \quad \text{i.e.,} \quad \epsilon^{\text{eff}} = \frac{\partial^2 \mathbf{G}^{\text{eff}}(\bar{\mathbf{e}})}{\partial \bar{\mathbf{e}} \partial \bar{\mathbf{e}}}. \quad (4.3.16)$$

For ideal dielectric materials, the electrostrictive stress due to the electric field defined previously in equations (4.2.29) would be written as

$$\boldsymbol{\sigma}_{\text{elect}} = \mathbb{A}_{\#}(\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}), \quad (4.3.17)$$

where the fourth order tensor $\mathbb{A}_{\#}$ is given by

$$\mathbb{A}_{\#}(\nabla \xi \otimes \nabla \xi) = \epsilon_{\#} \nabla \xi \otimes \nabla \xi - \frac{\epsilon_{\#}}{2} |\nabla \xi|^2. \quad (4.3.18)$$

By (4.3.9), (4.3.15), (4.3.18) and (4.3.12), we get

$$\text{div} \boldsymbol{\sigma}_{\text{elect}} = 0 \quad \text{and} \quad \text{div} \boldsymbol{\sigma}_{\text{mag}} = 0. \quad (4.3.19)$$

In other words, the electrostrictive and the magnetostrictive stresses are equivalent to an interfacial traction on the surface $\partial\Omega_0$. For a prescribed average strain $\bar{\mathbf{E}}$, we call $\bar{\mathcal{U}}_{\bar{\mathbf{E}}}$ the set of admissible displacement $\mathbf{u}$ with average strain $\bar{\mathbf{E}}$,

$$\mathcal{U}_{\bar{\mathbf{E}}} \equiv \{\mathbf{u} : \oint_Y \nabla \mathbf{u} = \bar{\mathbf{E}} \quad \text{and} \quad \nabla \mathbf{u} \text{ is } Y\text{-periodic}\}. \quad (4.3.20)$$

The actual local strain is uniquely determined by the following variation problem:

$$\Psi^{\text{eff}}(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}}) := \min\{F_2[\mathbf{u}, \bar{\mathbf{e}}, \bar{\mathbf{h}}]; \mathbf{u} \in \mathcal{U}_{\bar{\mathbf{E}}}\}, \quad (4.3.21)$$

where the energy $F_2[\mathbf{u}, \bar{\mathbf{e}}, \bar{\mathbf{h}}]$ can be rewritten as follows (equations (4.2.18) and (4.3.5)):

$$\begin{aligned} F_{(2)}[\mathbf{u}, \bar{\mathbf{e}}, \bar{\mathbf{h}}] = & \int_Y \frac{1}{2} \nabla \mathbf{u} \cdot \mathbb{C}_{\#} \nabla \mathbf{u} d\mathbf{x} + \\ & \int_{\partial\Omega_0} \frac{1}{2} [\nabla_s \mathbf{u} \cdot \mathbb{C}_s \nabla_s \mathbf{u} - \frac{2\gamma}{R_0} (\mathbf{u} \otimes \mathbf{n}) \cdot \mathbb{T} \nabla_s \mathbf{u}] d\mathbf{x} + \int_Y \nabla \mathbf{u} \cdot (\boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) d\mathbf{x}. \end{aligned} \quad (4.3.22)$$

The last integral comes from the electrostrictive and magnetostrictive stresses due to the existence of electric and magnetic field. We are interested in the effective electrostrictive and magnetostrictive properties of the composite and the effect of the surface tension γ on both of them. For that purpose, we follow the classic homogenization method. We define the effective stiffness tensor $\mathbb{C}^{\text{eff}}$, the effective electrostriction tensor $\mathbb{A}^{\text{eff}}$ and the effective magnetostriction tensor $\mathbb{B}^{\text{eff}}$ by equating the energy density of the composite to the energy density of the homogenized bulk material with the same average strain, electric field and magnetic field

$$\begin{aligned} \Psi_2^{\text{eff}}(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}}) = & \Psi_2^{\text{eff}}(0, 0, 0) + \sigma^0 \cdot \bar{\mathbf{E}} + \\ & \frac{1}{2} \bar{\mathbf{E}} \cdot \mathbb{C}^{\text{eff}} \bar{\mathbf{E}} + \bar{\mathbf{E}} \cdot \mathbb{A}^{\text{eff}} (\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) + \bar{\mathbf{E}} \cdot \mathbb{B}^{\text{eff}} (\bar{\mathbf{h}} \otimes \bar{\mathbf{h}}) + \dots \end{aligned} \quad (4.3.23)$$

We define the effective tensors $\mathbb{C}^{\text{eff}}$, $\mathbb{A}^{\text{eff}}$ and $\mathbb{B}^{\text{eff}}$ as

$$\begin{aligned}\mathbb{C}^{\text{eff}} &= \frac{\partial^2 \Psi_2^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{E}}} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}})=0} \\ \mathbb{A}^{\text{eff}} &= \frac{1}{2} \frac{\partial^3 \Psi_2^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{e}} \partial \bar{\mathbf{e}}} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}})=0} \\ \mathbb{B}^{\text{eff}} &= \frac{1}{2} \frac{\partial^3 \Psi_2^{\text{eff}}}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{h}} \partial \bar{\mathbf{h}}} \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}})=0}.\end{aligned}\tag{4.3.24}$$

The effective stiffness tensor and the effective electrostrictive tensor have been calculated in a previous work [78]. The effective magnetostrictive tensor can be determined by

$$\mathbb{B}^{\text{eff}} = \frac{1}{2} \frac{\partial^3}{\partial \bar{\mathbf{E}} \partial \bar{\mathbf{h}} \partial \bar{\mathbf{h}}} F_2[\mathbf{u}_{\bar{\mathbf{E}}}, \bar{\mathbf{e}}, \bar{\mathbf{h}}] \Big|_{(\bar{\mathbf{E}}, \bar{\mathbf{e}}, \bar{\mathbf{h}})=0},\tag{4.3.25}$$

where $\mathbf{u}_{\bar{\mathbf{E}}} \in \mathcal{U}_{\bar{\mathbf{E}}}$ is a minimizer of (4.3.21) and satisfy the following Euler-Lagrange equations:

$$\begin{cases} \operatorname{div}(\mathbb{C}^{\text{ex}} \nabla \mathbf{u} + \boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) = \operatorname{div}(\mathbb{C}^{\text{in}} \nabla \mathbf{u}) = 0 & \text{in } Y \setminus \Omega_0 \\ \operatorname{div}(\mathbb{C}^{\text{in}} \nabla \mathbf{u} + \boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) = \operatorname{div}(\mathbb{C}^{\text{ex}} \nabla \mathbf{u}) = 0 & \text{in } \Omega_0 \\ \operatorname{div}_s(\mathbb{C}_s \nabla_s \mathbf{u}) = \mathbf{t}_s & \text{on } \partial\Omega_0. \end{cases}\tag{4.3.26}$$

The surface traction $\mathbf{t}_s : \partial\Omega_0 \rightarrow \mathbb{R}$ due to the bulk elastic and magnetic stresses is given by

$$\begin{aligned}\mathbf{t}_s &= \{ [\mathbb{C}^{\text{in}} \nabla \mathbf{u} + \mathbb{A}_{\#}(\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}) + \mathbb{B}_{\#}(\nabla \zeta_{\bar{\mathbf{h}}} \otimes \nabla \zeta_{\bar{\mathbf{h}}})] \Big|_{\partial\Omega_0^-} \\ &\quad - [\mathbb{C}^{\text{ex}} \nabla \mathbf{u} + \mathbb{A}_{\#}(\nabla \xi_{\bar{\mathbf{e}}} \otimes \nabla \xi_{\bar{\mathbf{e}}}) + \mathbb{B}_{\#}(\nabla \zeta_{\bar{\mathbf{h}}} \otimes \nabla \zeta_{\bar{\mathbf{h}}})] \Big|_{\partial\Omega_0^+} \} \mathbf{n}.\end{aligned}\tag{4.3.27}$$

For isotropic composites, the electrostrictive and magnetostrictive tensors $\mathbb{A}^{\text{eff}}$, and $\mathbb{B}^{\text{eff}}$ have to be of the same form as an isotropic elasticity tensor and can be written as

$$\begin{aligned}\mathbb{A}^{\text{eff}} &= m_K^{\text{eff}} \mathcal{K} + m_J^{\text{eff}} \mathcal{J} \quad ; \quad \mathbb{B}^{\text{eff}} = n_K^{\text{eff}} \mathcal{K} + n_J^{\text{eff}} \mathcal{J} \\ \mathcal{K}_{ijkl} &= \frac{1}{2} [\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl}] \quad \text{and} \quad \mathcal{J}_{ijkl} = \frac{1}{3} \delta_{ij} \delta_{kl},\end{aligned}\tag{4.3.28}$$

where $m_K^{\text{eff}}, m_J^{\text{eff}}, n_K^{\text{eff}}, n_J^{\text{eff}}$ are materials constant in analogy with shear and bulk moduli of an isotropic elastic material. If $\mathbb{A}^{\text{eff}}$ and $\mathbb{B}^{\text{eff}}$ are isotropic, we find the relations between the components of the tensor $\mathbb{A}^{\text{eff}}$ and $\mathbb{B}^{\text{eff}}$ and the constants $m_K^{\text{eff}}, m_J^{\text{eff}}$ and $n_K^{\text{eff}}, n_J^{\text{eff}}$

respectively satisfy

$$\begin{aligned}\mathbb{A}_{1133}^{\text{eff}} = \mathbb{A}_{2233}^{\text{eff}} &= -\frac{1}{3}m_K^{\text{eff}} + \frac{1}{3}m_J^{\text{eff}} & ; & \quad \mathbb{B}_{1133}^{\text{eff}} = \mathbb{B}_{2233}^{\text{eff}} = -\frac{1}{3}n_K^{\text{eff}} + \frac{1}{3}n_J^{\text{eff}} \\ \mathbb{A}_{3333}^{\text{eff}} &= \frac{2}{3}m_K^{\text{eff}} + \frac{1}{3}m_J^{\text{eff}} & ; & \quad \mathbb{B}_{3333}^{\text{eff}} = \frac{2}{3}n_K^{\text{eff}} + \frac{1}{3}n_J^{\text{eff}}.\end{aligned}\tag{4.3.29}$$

4.3.3 Exact solution for a coated sphere with surface energy

For exact solutions, we consider the special microstructure of Hashin's coated spheres. Let $\mathbf{u} \in \mathcal{U}_{\bar{\mathbf{E}}}$ be a trial solution satisfying (4.3.26)_{1,2} (but not necessarily (4.3.26)₃), $\mathbf{u}'$ be the "astray" displacement

$$\mathbf{u}' = \mathbf{u} - \bar{\mathbf{u}}, \quad \bar{\mathbf{u}} = \bar{\mathbf{E}}\mathbf{x}.\tag{4.3.30}$$

We remark that $\mathbf{u}'$ itself is Y -periodic. Upon specifying the boundary displacement

$$\mathbf{u}' = \mathbf{u}'_b \quad \text{on } \partial\Omega_0,$$

the differential equations (4.3.26)_{1,2} uniquely determine the "astray" displacement $\mathbf{u}'$ in both the exterior region $Y \setminus \Omega_0$ and the interior region Ω_0 . Since the inclusion Ω_0 is a sphere of radius R_0 , we employ spherical coordinates (r, θ, ϕ) and vector spherical harmonics, and write the boundary value of $\mathbf{u}'$ on $\partial\Omega_0$ as

$$\mathbf{u}'_b(\theta) = \sum_{l=0}^{\infty} [X'_l \mathbf{Y}_l(\theta) + \Theta'_l \mathbf{\Psi}_l(\theta)],\tag{4.3.31}$$

where the vectorial axis-symmetric spherical harmonics are defined as

$$\mathbf{Y}_l(\theta) = Y_l(\theta)\mathbf{e}_r, \quad \mathbf{\Psi}_l = r\nabla Y_l = \mathbf{e}_\theta \frac{d}{d\theta} Y_l(\theta),\tag{4.3.32}$$

and $(\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\phi)$ are the unit basis vectors associated with spherical coordinates (r, θ, ϕ) .

We combine the first two integral terms in the free energy integral (4.3.22) together and

denote that by $F_1[\mathbf{u}]$. This can be rewritten as [78]

$$F_1[\mathbf{u}] = \frac{1}{2} \left[v_f \bar{\mathbf{E}} \cdot \mathbb{C}^{\text{in}} \bar{\mathbf{E}} + (1 - v_f) \bar{\mathbf{E}} \cdot \mathbb{C}^{\text{ex}} \bar{\mathbf{E}} \right] + \sum_{l=0,2} \frac{1}{2} \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix} \cdot \mathbf{S}_l \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix} + Q_1(X'_l, \Theta'_l), \quad (4.3.33)$$

where

$$Q_1(X'_l, \Theta'_l) = \sum_{l=0,2} \left\{ \frac{1}{2} \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \cdot (\mathbf{D}_l^{\text{in}} + \mathbf{D}_l^{\text{ex}} + \mathbf{S}_l) \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} + \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \cdot (\mathbf{S}_l + \mathbf{B}_l) \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix} \right\}. \quad (4.3.34)$$

The matrices $\mathbf{D}_l^{\text{in}}, \mathbf{D}_l^{\text{ex}}, \mathbf{S}_l, \mathbf{B}_l$ that appear in the above expression are defined as follows:

$$\begin{aligned} \mathbf{D}_0^{\text{in}} &= R_0 \kappa^{\text{in}} \begin{bmatrix} 3 & 0 \\ 0 & 0 \end{bmatrix}, \quad \mathbf{D}_0^{\text{ex}} = \frac{R_0(4\mu^{\text{ex}} + 3\kappa^{\text{ex}} v_f)}{1 - v_f} \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}, \\ \mathbf{D}_2^{\text{in}} &= \frac{R_0 \mu^{\text{in}}}{6\kappa^{\text{in}} + 17\mu^{\text{in}}} \begin{bmatrix} 39\kappa^{\text{in}} + 16\mu^{\text{in}} & 18(-3\kappa^{\text{in}} + 2\mu^{\text{in}}) \\ 18(-3\kappa^{\text{in}} + 2\mu^{\text{in}}) & 12(15\kappa^{\text{in}} + 11\mu^{\text{in}}) \end{bmatrix}, \\ \mathbf{D}_2^{\text{ex}} &= \frac{2\mu^{\text{ex}} R_0}{3(1 - v_f)(\kappa^{\text{ex}} + 2\mu^{\text{ex}})} \begin{bmatrix} 9\kappa^{\text{ex}} + 16\mu^{\text{ex}} - 6v_f(\kappa^{\text{ex}} + 2\mu^{\text{ex}}) & 3(\kappa^{\text{ex}}(6v_f - 3) + 4\mu^{\text{ex}}(3v_f - 2)) \\ 3(\kappa^{\text{ex}}(6v_f - 3) + 4\mu^{\text{ex}}(3v_f - 2)) & 9(5\kappa^{\text{ex}} + 8\mu^{\text{ex}} - 2v_f(\kappa^{\text{ex}} + 2\mu^{\text{ex}})) \end{bmatrix}, \\ \mathbf{B}_0 &= 3(\kappa^{\text{in}} - \kappa^{\text{ex}}) R_0 \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}, \quad \mathbf{B}_2 = -2R_0(\mu^{\text{ex}} - \mu^{\text{in}}) \begin{bmatrix} 1 & 0 \\ 0 & 6 \end{bmatrix}, \quad \mathbf{S}_l = \gamma \begin{bmatrix} l(l+1) - 2 & 0 \\ 0 & 0 \end{bmatrix}. \end{aligned} \quad (4.3.35)$$

Also, we write the displacement $\bar{\mathbf{u}} = \bar{\mathbf{E}}\mathbf{x} = (ax_1, ax_2, bx_3)$ on $\partial\Omega_0$ as

$$\bar{\mathbf{u}} = \bar{X}_0 \mathbf{Y}_0 + \bar{X}_2 \mathbf{Y}_2 + \bar{\Theta}_2 \mathbf{\Psi}_2, \quad (4.3.36)$$

where

$$\bar{X}_0 = \frac{2}{3}R_0\pi^{1/2}(2a+b), \quad \bar{X}_2 = -\frac{4}{15}R_0(5\pi)^{1/2}(a-b) \quad \bar{\Theta}_2 = -\frac{2}{15}R_0(5\pi)^{1/2}(a-b).$$

The calculations above have been reported in details in a previous work [78].

For a coated sphere, the conditions $-f_Y \nabla \xi = \bar{\mathbf{e}}$ and $-f_Y \nabla \zeta = \bar{\mathbf{h}}$ should be respectively replaced by

$$\begin{aligned} \int_{\{r=R\}} \xi \mathbf{e}_r &= \frac{4\pi R^3}{3} \bar{\mathbf{e}}. \\ \int_{r=R} \zeta \mathbf{e}_r &= \frac{4\pi R^3}{3} \bar{\mathbf{e}}. \end{aligned} \tag{4.3.37}$$

If the average electric field $\bar{\mathbf{e}} = e_0 \mathbf{e}_3$, the solution to the electrostatic problem (4.3.15) and (4.3.37)₂ is well-known and can be found in the textbook

$$\xi_{\bar{\mathbf{e}}} = \begin{cases} -c_0^{\text{in}} e_0 r \cos \theta & \text{if } r \leq R_0, \\ -c_1^{\text{ex}} e_0 r \cos \theta - c_2^{\text{ex}} e_0 \frac{\cos \theta}{r^2} & \text{if } R_0 < r < R, \end{cases} \tag{4.3.38}$$

where

$$\begin{aligned} c_0^{\text{in}} &= \frac{3\epsilon^{\text{ex}}}{(v_f + 2)\epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}}}, \\ c_1^{\text{ex}} &= \frac{2\epsilon^{\text{ex}} + \epsilon^{\text{in}}}{(v_f + 2)\epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}}} \quad \text{and} \\ c_2^{\text{ex}} &= \frac{R_0^3(\epsilon^{\text{ex}} - \epsilon^{\text{in}})}{(v_f + 2)\epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}}}. \end{aligned}$$

Similarly, if the average magnetic field $\bar{\mathbf{h}} = h_0 \mathbf{e}_3$, the solution to the magnetostatic problem (4.3.9) and (4.3.37)₁ is also well-known and is given by

$$\zeta_{\bar{\mathbf{h}}} = \begin{cases} -k_0^{\text{in}} h_0 r \cos \theta & \text{if } r \leq R_0 \\ -k_1^{\text{ex}} h_0 r \cos \theta - k_2^{\text{ex}} h_0 \frac{\cos \theta}{r^2} & \text{if } R_0 < r < R, \end{cases} \tag{4.3.39}$$

where

$$\begin{aligned} k_0^{\text{in}} &= \frac{3\mu_0}{\mu_0(v_f + 2) + \mu^{\text{in}} - \mu^{\text{in}}v_f}, \\ k_1^{\text{ex}} &= \frac{(2\mu_0 + \mu^{\text{in}})}{\mu^{\text{in}} + \mu_0(v_f + 2) - \mu^{\text{in}}v_f} \quad \text{and} \\ k_2^{\text{ex}} &= \frac{R_0^3(\mu_0 - \mu^{\text{in}})}{\mu^{\text{in}} + \mu_0(v_f + 2) - \mu^{\text{in}}v_f}. \end{aligned}$$

Immediately, from the definitions (4.3.13) and (4.3.10), we find the effective permittivity and permeability of the composite

$$\epsilon^{\text{eff}} = \epsilon^{\text{ex}} + \frac{3v_f\epsilon^{\text{ex}}(-\epsilon^{\text{ex}} + \epsilon^{\text{in}})}{(2 + v_f)\epsilon^{\text{ex}} + \epsilon^{\text{in}} - v_f\epsilon^{\text{in}}} \quad (4.3.40)$$

and

$$\mu^{\text{eff}} = \mu_0 + \frac{3v_f\mu_0(-\mu_0 + \mu^{\text{in}})}{(2 + v_f)\mu_0 + \mu^{\text{in}} - v_f\mu^{\text{in}}}. \quad (4.3.41)$$

To solve the minimization problem (4.3.21), it is convenient to rewrite the last term in eq.(4.3.22) as a surface integral. First, we notice that

$$\begin{aligned} \int_Y \nabla \mathbf{u} \cdot (\boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) &= \int_Y (\bar{\mathbf{E}} + \nabla \mathbf{u}') \cdot (\boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) \\ &= \int_Y \bar{\mathbf{E}} \cdot \boldsymbol{\sigma}_{\text{elect}} + \int_Y \bar{\mathbf{E}} \cdot \boldsymbol{\sigma}_{\text{mag}} - \int_{\partial\Omega_0} \mathbf{u}' \cdot \llbracket \boldsymbol{\sigma}_{\text{elect}} \rrbracket \mathbf{n} - \int_{\partial\Omega_0} \mathbf{u}' \cdot \llbracket \boldsymbol{\sigma}_{\text{mag}} \rrbracket \mathbf{n}. \end{aligned} \quad (4.3.42)$$

From (??), we find that

$$\begin{aligned} \int_Y \bar{\mathbf{E}} \cdot \boldsymbol{\sigma}_{\text{elect}} &= \frac{2}{3}\pi R_0^3(e_0)^2\epsilon^{\text{in}}(c_0^{\text{in}})^2(b - 2a) \\ &+ \frac{2\pi e_0^2 R_0^3(v_f - 1)\epsilon^{\text{ex}}}{15v_f((v_f + 2)\epsilon^{\text{ex}} - v_f\epsilon^{\text{in}} + \epsilon^{\text{in}})^2} \left(2(\epsilon^{\text{ex}})^2(4a(v_f + 5) + b(v_f - 10)) \right. \\ &\left. - 4\epsilon^{\text{ex}}\epsilon^{\text{in}}(2a(2v_f - 5) + b(v_f + 5)) + (\epsilon^{\text{in}})^2(2a(4v_f + 5) + b(2v_f - 5)) \right). \end{aligned} \quad (4.3.43)$$

Similarly, we also find that

$$\begin{aligned}
\int_Y \bar{\mathbf{E}} \cdot \boldsymbol{\sigma}_{\text{mag}} &= \frac{2}{3} \pi R_0^3 (h_0)^2 \mu^{\text{in}} (k_0^{\text{in}})^2 (b - 2a) \\
&+ \frac{2\pi h_0^2 R_0^3 (v_f - 1) \mu_0}{15 v_f ((v_f + 2) \mu_0 - v_f \mu^{\text{in}} + \mu^{\text{in}})^2} \left(2(\mu_0)^2 (4a(v_f + 5) + b(v_f - 10)) \right. \\
&\left. - 4\mu_0 \mu^{\text{in}} (2a(2v_f - 5) + b(v_f + 5)) + (\mu^{\text{in}})^2 (2a(4v_f + 5) + b(2v_f - 5)) \right). \quad (4.3.44)
\end{aligned}$$

In addition, we obtain

$$\begin{aligned}
- \int_{\partial\Omega_0} \mathbf{u}' \cdot \llbracket \boldsymbol{\sigma}_{\text{elect}} \rrbracket \mathbf{n} &= \sum_{l=0,2} \left\{ \mathbf{b}_l \cdot \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \right\} \\
&\text{and} \quad (4.3.45)
\end{aligned}$$

$$- \int_{\partial\Omega_0} \mathbf{u}' \cdot \llbracket \boldsymbol{\sigma}_{\text{mag}} \rrbracket \mathbf{n} = \sum_{l=0,2} \left\{ \mathbf{d}_l \cdot \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \right\}$$

where

$$\begin{aligned}
\mathbf{b}_0 &= \left[\frac{3\sqrt{\pi} e_0^2 R_0^2 \epsilon^{\text{ex}} (2(\epsilon^{\text{ex}})^2 - \epsilon^{\text{ex}} \epsilon^{\text{in}} - (\epsilon^{\text{in}})^2)}{((v_f + 2) \epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}})^2}, 0 \right], & \mathbf{d}_0 &= \left[\frac{3\sqrt{\pi} h_0^2 R_0^2 \mu_0 (\mu_0 - \mu^{\text{in}}) (2\mu_0 + \mu^{\text{in}})}{((v_f + 2) \mu_0 - v_f \mu^{\text{in}} + \mu^{\text{in}})^2}, 0 \right], \\
\mathbf{b}_2 &= \left[-\frac{6\sqrt{\frac{\pi}{5}} e_0^2 R_0^2 \epsilon^{\text{ex}} (\epsilon^{\text{ex}} - \epsilon^{\text{in}})^2}{((v_f + 2) \epsilon^{\text{ex}} - v_f \epsilon^{\text{in}} + \epsilon^{\text{in}})^2}, 0 \right], & \mathbf{d}_2 &= \left[-\frac{6\sqrt{\frac{\pi}{5}} h_0^2 R_0^2 \mu_0 (\mu_0 - \mu^{\text{in}})^2}{((v_f + 2) \mu_0 - v_f \mu^{\text{in}} + \mu^{\text{in}})^2}, 0 \right].
\end{aligned}$$

Collecting terms in (4.3.22), by (4.3.33)-(4.3.36), and (4.3.42)-(4.3.45), we once again manage to write the total energy as a quadratic function of (X'_l, Θ'_l)

$$\begin{aligned}
F_2[\mathbf{u}; \bar{\mathbf{e}}] &= \frac{1}{2} \bar{\mathbf{E}} \cdot [v_f \mathbb{C}^{\text{in}} + (1 - v_f) \cdot \mathbb{C}^{\text{ex}}] \bar{\mathbf{E}} + \frac{1}{2} \gamma (-2\bar{X}_0^2 + 4\bar{X}_2^2) \\
&+ \bar{\mathbf{E}} \cdot \int_Y (\boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_{\text{mag}}) + Q_2(X'_l, \Theta'_l), \quad (4.3.46)
\end{aligned}$$

where

$$\begin{aligned}
Q_2(X'_l, \Theta'_l) &= Q_1(X'_l, \Theta'_l) + \sum_{l=0,2} \left\{ (\mathbf{b}_l + \mathbf{d}_l) \cdot \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \right\} \\
&= \frac{1}{2} \sum_{l=0,2} \left\{ \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \cdot (\mathbf{D}_l^{\text{in}} + \mathbf{D}_l^{\text{ex}} + \mathbf{S}_l) \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} + \begin{bmatrix} X'_l \\ \Theta'_l \end{bmatrix} \cdot \left\{ (\mathbf{S}_l + \mathbf{B}_l) \begin{bmatrix} \bar{X}_l \\ \bar{\Theta}_l \end{bmatrix} + \mathbf{b}_l + \mathbf{d}_l \right\} \right\}. \quad (4.3.47)
\end{aligned}$$

From the original variation problem (4.3.21), the equilibrium X'_0, X'_2, Θ'_2 can be determined by minimizing Q_2

$$Q_2^*(a, b) := \min_{X'_0, X'_2, \Theta'_2} Q_2(X'_0, X'_2, \Theta'_2). \quad (4.3.48)$$

Being a quadratic minimization problem, we immediately find the minimizing (X'_0, X'_2, Θ'_2) . Inserting the solution back into (4.3.48), we find the minimum free energy as a function of (a, b) . Then, using eq.(4.3.24), we can find the effective electrostrictive and magnetostrictive tensors. In particular, we can explicitly calculate the components $\mathbb{A}_{1133}^{\text{eff}}, \mathbb{A}_{3333}^{\text{eff}}, \mathbb{B}_{1133}^{\text{eff}}$ and $\mathbb{B}_{3333}^{\text{eff}}$. The calculations for the effective electrostrictive tensor $\mathbb{A}^{\text{eff}}$ and the materials constants $m_K^{\text{eff}}, m_J^{\text{eff}}$ have been reported in [78] and will be used here for the sake of completeness. They are given by

$$\begin{aligned} m_K^{\text{eff}} &= \epsilon^{\text{ex}} + \Delta\epsilon^2 \frac{1}{15v_f} \left(\frac{5-2v_f}{\epsilon^{\text{ex}}} + \frac{15}{\epsilon^{\text{in}} - \epsilon^{\text{ex}}} \right) \\ &\quad + \Delta\epsilon^2 \Delta G \left(-\frac{1}{5v_f^2 \epsilon^{\text{ex}} \left(-G^{\text{ex}} + G^{\text{in}} + \frac{4\gamma}{R_0} \frac{6\kappa^{\text{in}} G^{\text{ex}} + 15\kappa^{\text{in}} G^{\text{in}} + 17G^{\text{ex}} G^{\text{in}} + 11(G^{\text{in}})^2}{I_2} \right)} \right), \quad (4.3.49) \\ m_J^{\text{eff}} &= -\frac{\epsilon^{\text{ex}}}{2} - \frac{1}{2} \Delta\epsilon - \Delta\epsilon^2 \frac{(-1+v_f)(2\epsilon^{\text{ex}} + \epsilon^{\text{in}}) \left(\frac{2\gamma}{R_0} + 3(\kappa^{\text{ex}} - \kappa^{\text{in}}) \right)}{2v_f \epsilon^{\text{ex}} (\epsilon^{\text{ex}} - \epsilon^{\text{in}}) \left(\frac{2(-1+v_f)\gamma}{R_0} + 3v_f(\kappa^{\text{ex}} - \kappa^{\text{in}}) + 3\kappa^{\text{in}} + 4\mu^{\text{ex}} \right)}. \end{aligned}$$

On the other hand, the materials constants corresponding to the magnetostrictive tensor are given by

$$\begin{aligned} n_K^{\text{eff}} &= \mu_0 + \Delta\mu^2 \left(\frac{5-2v_f + \frac{15\mu_0}{-\mu_0 + \mu^{\text{in}}}}{15v_f \mu_0} \right) \\ &\quad + \Delta\mu^2 \Delta G \left(-\frac{1}{5v_f^2 \left(-G^{\text{ex}} + G^{\text{in}} + \frac{4\gamma(17G^{\text{ex}} G^{\text{in}} + 11(G^{\text{in}})^2 + 6G^{\text{ex}} \kappa^{\text{in}} + 15G^{\text{in}} \kappa^{\text{in}})}{R_0(4G^{\text{in}}(34G^{\text{ex}} + G^{\text{in}}) + (48G^{\text{ex}} + 57G^{\text{in}})\kappa^{\text{in}})} \right)} \mu_0 \right), \\ n_J^{\text{eff}} &= -\frac{\mu_0}{2} - \frac{1}{2} \Delta\mu + \Delta\mu^2 \left(\frac{(1-v_f) \left(\frac{2\gamma}{R_0} + 3(\kappa^{\text{ex}} - \kappa^{\text{in}}) \right) (2\mu_0 + \mu^{\text{in}})}{2v_f \left(4G^{\text{ex}} + \frac{2(-1+v_f)\gamma}{R_0} + 3(v_f \kappa^{\text{ex}} + \kappa^{\text{in}} - v_f \kappa^{\text{in}}) \right) \mu_0 (\mu_0 - \mu^{\text{in}})} \right). \quad (4.3.50) \end{aligned}$$

We define $\Delta G = G^{\text{eff}} - G^{\text{ex}}$, $\Delta\epsilon = \epsilon^{\text{eff}} - \epsilon^{\text{ex}}$, and $\Delta\mu = \mu^{\text{eff}} - \mu^{\text{ex}}$.

The above results have few limiting cases:

1. Dilute volume fraction of particles:

$$m_K^{\text{eff}} = \epsilon^{\text{ex}} + \frac{3v_f \epsilon^{\text{ex}}(-\epsilon^{\text{ex}} + \epsilon^{\text{in}})}{2\epsilon^{\text{ex}} + \epsilon^{\text{in}}} - \left(9I_2 v_f \epsilon^{\text{ex}} (\epsilon^{\text{ex}} - \epsilon^{\text{in}})^2 G^{\text{ex}} (3\kappa^{\text{ex}} + 4G^{\text{ex}}) \right) /$$

$$(2\epsilon^{\text{ex}} + \epsilon^{\text{in}})^2 \left(I_2 (G^{\text{ex}} (9\kappa^{\text{ex}} + 8G^{\text{ex}}) + 6(\kappa^{\text{ex}} + 2G^{\text{ex}}) G^{\text{in}}) + \frac{12\gamma}{R_0} (30\kappa^{\text{ex}} \kappa^{\text{in}} (G^{\text{ex}} + G^{\text{in}}) \right.$$

$$\left. + 12\kappa^{\text{in}} G^{\text{ex}} (4G^{\text{ex}} + 5G^{\text{in}}) + 4G^{\text{ex}} G^{\text{in}} (34G^{\text{ex}} + 11G^{\text{in}}) + \kappa^{\text{ex}} G^{\text{in}} (85G^{\text{ex}} + 22G^{\text{in}}) \right),$$

$$m_J^{\text{eff}} = \frac{1}{2} \epsilon^{\text{ex}} \left(-1 - \frac{3v_f (\epsilon^{\text{ex}} - \epsilon^{\text{in}}) \left(\frac{4\gamma}{R_0} + 9\kappa^{\text{ex}} - 6\kappa^{\text{in}} + 4G^{\text{ex}} \right)}{(2\epsilon^{\text{ex}} + \epsilon^{\text{in}}) \left(\frac{2\gamma}{R_0} - 3\kappa^{\text{in}} - 4G^{\text{ex}} \right)} \right),$$

$$n_K^{\text{eff}} = \mu_0 + \frac{3v_f \mu_0 (-\mu_0 + \mu^{\text{in}})}{2\mu_0 + \mu^{\text{in}}} - \left(9I_2 v_f \mu_0 (\mu_0 - \mu^{\text{in}})^2 G^{\text{ex}} (3\kappa^{\text{ex}} + 4G^{\text{ex}}) \right) /$$

$$(2\mu_0 + \mu^{\text{in}})^2 \left(I_2 (G^{\text{ex}} (9\kappa^{\text{ex}} + 8G^{\text{ex}}) + 6(\kappa^{\text{ex}} + 2G^{\text{ex}}) G^{\text{in}}) + \frac{12\gamma}{R_0} (30\kappa^{\text{ex}} \kappa^{\text{in}} (G^{\text{ex}} + G^{\text{in}}) \right.$$

$$\left. + 12\kappa^{\text{in}} G^{\text{ex}} (4G^{\text{ex}} + 5G^{\text{in}}) + 4G^{\text{ex}} G^{\text{in}} (34G^{\text{ex}} + 11G^{\text{in}}) + \kappa^{\text{ex}} G^{\text{in}} (85G^{\text{ex}} + 22G^{\text{in}}) \right),$$

$$n_J^{\text{eff}} = \frac{1}{2} \mu_0 \left(-1 - \frac{3v_f (\mu_0 - \mu^{\text{in}}) \left(\frac{4\gamma}{R_0} + 9\kappa^{\text{ex}} - 6\kappa^{\text{in}} + 4G^{\text{ex}} \right)}{(2\mu^{\text{ex}} + \mu^{\text{in}}) \left(\frac{2\gamma}{R_0} - 3\kappa^{\text{in}} - 4G^{\text{ex}} \right)} \right). \quad (4.3.51)$$

2. Rigid particles with infinite permeability: In the limit of rigid infinite-permeability particles when $G^{\text{in}}, \kappa^{\text{in}} \rightarrow +\infty$ and $\mu^{\text{in}} \rightarrow \infty$

$$n_K^{\text{eff}} = \mu_0 + \frac{3(5 - 2v_f)v_f \mu_0}{5(1 - v_f)^2},$$

$$n_J^{\text{eff}} = -\frac{\mu_0}{2} + \frac{3v_f(2 + v_f)\mu_0}{2(1 - v_f)^2}. \quad (4.3.52)$$

We need to note that, even if the matrix is incompressible ($\kappa^{\text{ex}} \rightarrow +\infty$), the results will not change in this limit.

3. Liquid-like particles with infinite permeability: $G^{\text{in}} \rightarrow 0, \kappa^{\text{in}} \rightarrow +\infty$ and $\mu^{\text{in}} \rightarrow +\infty$

$$n_K^{\text{eff}} = \mu_0 + \frac{3v_f \mu_0 \left(-\frac{3\gamma}{R_0} (-5 + 2v_f) ((-5 + 2v_f)\kappa^{\text{ex}} + 4(-2 + v_f)G^{\text{ex}}) - 8(1 - v_f)G^{\text{ex}} (-5G^{\text{ex}} + 3v_f(\kappa^{\text{ex}} + 2G^{\text{ex}})) \right)}{5(-1 + v_f)^2 \left(\frac{3\gamma}{R_0} ((-5 + 2v_f)\kappa^{\text{ex}} + 4(-2 + v_f)G^{\text{ex}}) - 2G^{\text{ex}} ((9 + 6v_f)\kappa^{\text{ex}} + 4(2 + 3v_f)G^{\text{ex}}) \right)},$$

$$n_J^{\text{eff}} = -\frac{\mu_0}{2} + \frac{3v_f(2 + v_f)\mu_0}{2(1 - v_f)^2}. \quad (4.3.53)$$

In addition, if the matrix is incompressible ($\kappa^{\text{ex}} \rightarrow +\infty$), then the above equations reduce to

$$\begin{aligned} n_K^{\text{eff}} &= \mu_0 - \frac{3v_f\mu_0 \left(\frac{\gamma}{R_0}(5-2v_f)^2 - 8(-1+v_f)v_f G^{\text{ex}} \right)}{5(-1+v_f)^2 \left(\frac{\gamma}{R_0}(-5+2v_f) - 2(3+2v_f)G^{\text{ex}} \right)}, \\ n_J^{\text{eff}} &= -\frac{\mu_0}{2} + \frac{3v_f(2+v_f)\mu_0}{2(1-v_f)^2}. \end{aligned} \quad (4.3.54)$$

4.4 Results and Discussion

In this section, we would like to measure the magnetostriction deformation that the body will undergo when subjected to an magnetic field $\bar{\mathbf{h}}$. From (4.3.26)_{1,2}, the stress is expressed as

$$\bar{\boldsymbol{\sigma}} = \mathbb{C}^{\text{eff}} \bar{\mathbf{E}} + \mathbb{A}^{\text{eff}} (\bar{\mathbf{e}} \otimes \bar{\mathbf{e}}) + \mathbb{B}^{\text{eff}} (\bar{\mathbf{h}} \otimes \bar{\mathbf{h}}). \quad (4.4.1)$$

If the applied stress $\bar{\boldsymbol{\sigma}} = 0$, and in the absence of the electric voltage, then, from (4.4.1), the average deformation can be written as

$$\bar{\mathbf{E}} = -\frac{n_K^{\text{eff}}}{2G^{\text{eff}}} \bar{\mathbf{h}} \otimes \bar{\mathbf{h}} + \left(\frac{n_K^{\text{eff}}}{6G^{\text{eff}}} - \frac{n_J^{\text{eff}}}{9\kappa^{\text{eff}}} \right) (\bar{\mathbf{h}} \cdot \bar{\mathbf{h}}) \mathbf{I}. \quad (4.4.2)$$

In order to measure the magnetostriction experimentally, people consider a dielectric thin film as sketched in 4.4.1. The thin film is subject to an external magnetic field $\bar{\mathbf{h}} = h_0 \mathbf{k}$. Note that in this case, we assume small deformations and won't distinguish between the reference and the deformed configuration. The deformation takes the following diagonal form

$$\bar{\mathbf{E}} = \bar{E}_{11} \mathbf{i} \otimes \mathbf{i} + \bar{E}_{22} \mathbf{j} \otimes \mathbf{j} + \bar{E}_{33} \mathbf{k} \otimes \mathbf{k}, \quad (4.4.3)$$

where

$$\bar{E}_{11} = \bar{E}_{22} = \left(\frac{n_K^{\text{eff}}}{6G^{\text{eff}}} - \frac{n_J^{\text{eff}}}{9\kappa^{\text{eff}}} \right) h_0^2 \quad \bar{E}_{33} = -\left(\frac{n_K^{\text{eff}}}{3G^{\text{eff}}} + \frac{n_J^{\text{eff}}}{9\kappa^{\text{eff}}} \right) h_0^2. \quad (4.4.4)$$

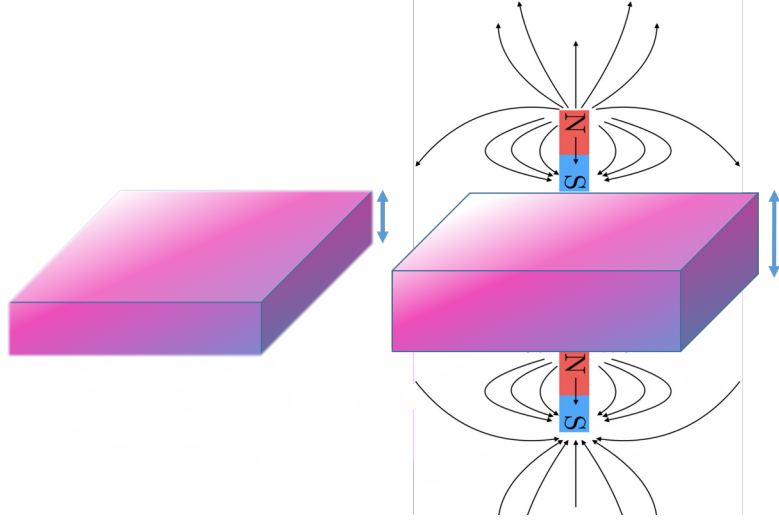


Figure 4.4.1: Schematic of magnetostriction under the application of a uniaxial magnetic field.

and

$$E_{11}^{\text{ex}} = E_{22}^{\text{ex}} = \left(\frac{\mu_0}{6G^{\text{ex}}} + \frac{\mu_0}{18\kappa^{\text{ex}}} \right) h_0^2 \quad E_{33}^{\text{ex}} = - \left(\frac{\mu_0}{3G^{\text{ex}}} - \frac{\mu_0}{18\kappa^{\text{ex}}} \right) h_0^2. \quad (4.4.5)$$

The ratios $\frac{\bar{E}_{11}}{E_{11}^{\text{ex}}}$ and $\frac{\bar{E}_{33}}{E_{33}^{\text{ex}}}$ will provide us with more understanding of the deformation provoked by the magnetostriction as we keep on adding more particles into the thin dielectric film. It will also reveal the impact of surface effect on the deformation if we deal with liquid like particles.

We start by investigating the behavior of the composite made of a soft dielectric film filled with liquid like magnetic particles under the application of an external magnetic field only. From our knowledge of the Maxwell's stress effect, we expect the composite to show some deformation in the presence of a magnetic field. We confirm that in Fig.4.4.2. The electric field is assumed to be zero and we consider several values of biaxial loads. We note from the graphs that the deformation increases as the magnetic field increases. We also note

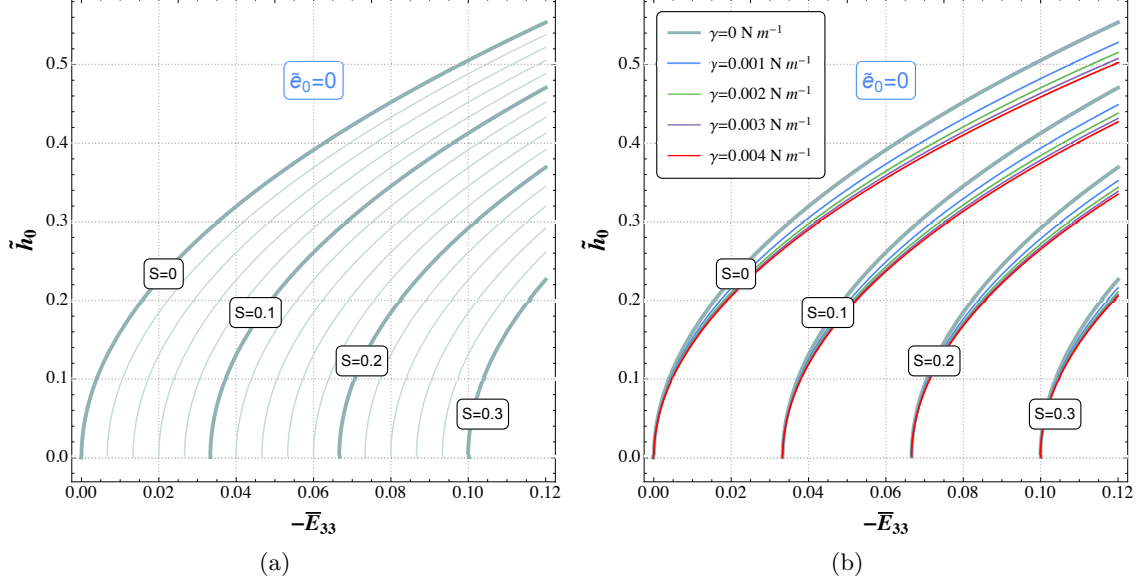


Figure 4.4.2: Behavior of a dielectric film filled with liquid-like magnetic spherical particles subjected to zero electric field under various biaxial loads ($\kappa^{\text{ex}} = 10^3 G^{\text{ex}}$, $G^{\text{in}} = 10^{-2} G^{\text{ex}}$, $\kappa^{\text{in}} = 10^3 G^{\text{ex}}$, $\mu^{\text{in}} = 10\mu_0$, $G^{\text{ex}} = 3 \text{ MPa}$, $v_f = 0.2$).

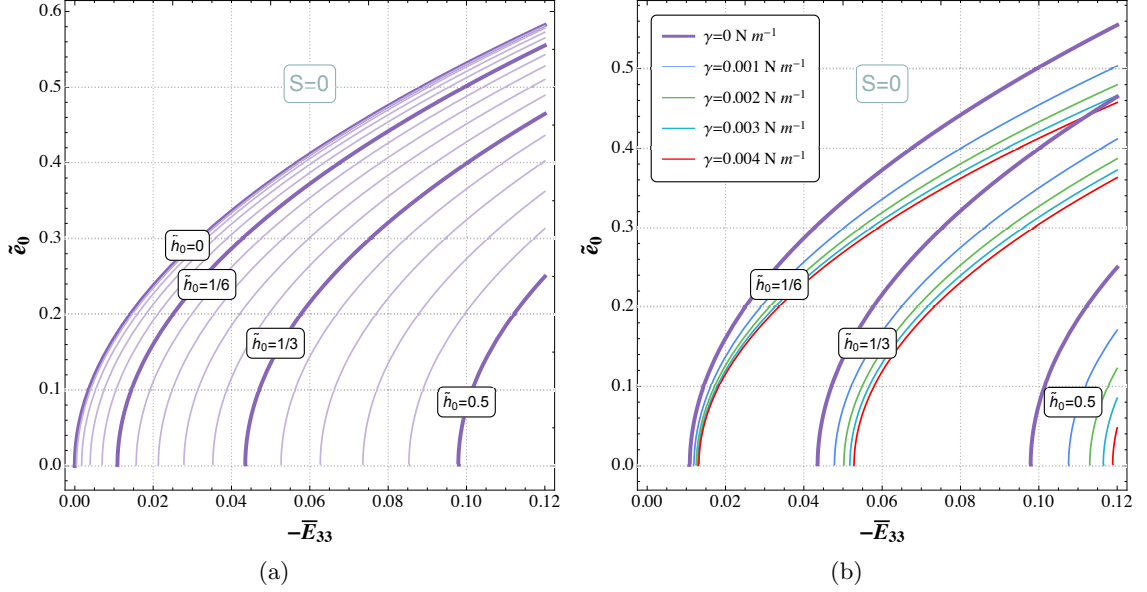


Figure 4.4.3: Behavior of a dielectric film filled with liquid-like spherical particles. ($\kappa^{\text{ex}} = 10^3 G^{\text{ex}}$, $G^{\text{in}} = 10^{-2} G^{\text{ex}}$, $\kappa^{\text{in}} = 10^3 G^{\text{ex}}$, $\epsilon^{\text{in}} = 10^2 \epsilon^{\text{ex}}$, $\mu^{\text{in}} = 10\mu_0$, $G^{\text{ex}} = 3 \text{ MPa}$, $\epsilon^{\text{ex}} = 3\epsilon_0$, $v_f = 0.2$)

that increasing the biaxial load increases the deformation of the composite. In Fig.4.2(a), surface tension is ignored, whereas in Fig.4.2(b) we graph the same results but we add the contribution of the surface tension. comparing both sides of Fig.4.4.2, we can conclude that

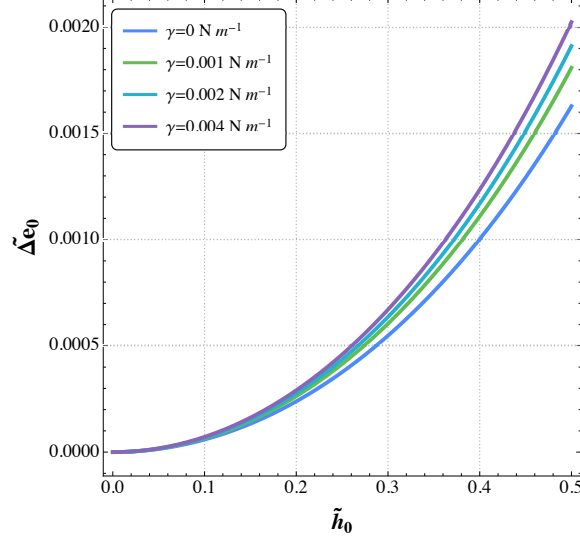


Figure 4.4.4: Variation of the electric field for various values of the applied magnetic field ($\kappa^{\text{ex}} = 10^3 G^{\text{ex}}, G^{\text{in}} = 10^{-2} G^{\text{ex}}, \kappa^{\text{in}} = 10^3 G^{\text{ex}}, \epsilon^{\text{in}} = 10^2 \epsilon^{\text{ex}}, \mu^{\text{in}} = 10 \mu_0, G^{\text{ex}} = 3 MPa, \epsilon^{\text{ex}} = 3 \epsilon_0, v_f = 0.2$)

surface tension makes the composite more susceptible to external magnetic field. Larger deformations are expected for the same $\bar{\mathbf{h}}_0$ when the surface tension is considered. If $\bar{\mathbf{h}}_0$ and S are kept constant, increasing γ results in larger deformation.

In Fig.4.4.3, we show the effect of the electric field on the composite for various values of the magnetic field and zero mechanical load. In Fig.4.3(a), we show the results without surface tension effect while we take that into consideration in Fig.4.3(b). We notice that both electric and magnetic fields deform the composite. If we look carefully at Fig.4.3(a), we notice that for $\gamma = 0.004 N.m^{-1}$, $\bar{\mathbf{h}}_0 = 1/6$ and $\bar{\mathbf{e}}_0 = 0.46$, the composite deforms similar to the case where $\bar{\mathbf{h}}_0 = 1/3$ and $\bar{\mathbf{e}} = 0.47$ in the absence of surface tension, i.e. the red line for $\bar{\mathbf{h}}_0 = 1/6$ is higher than the purple line for $\bar{\mathbf{h}}_0 = 1/3$ for $\bar{\mathbf{e}} > 0.4$. Thus, we conclude that the composite is capable of larger deformation for smaller electric and magnetic fields when capillary effects are considered. The implications of this result are crucial for designing soft materials that respond to smaller external fields with larger deformations.

In Fig.4.4.4, we show another significant result. We graph the change of the measured electric field as function of the applied magnetic field. It is clear from the graph that the output electric field changes a function of the magnetic field. This implies that it is possible

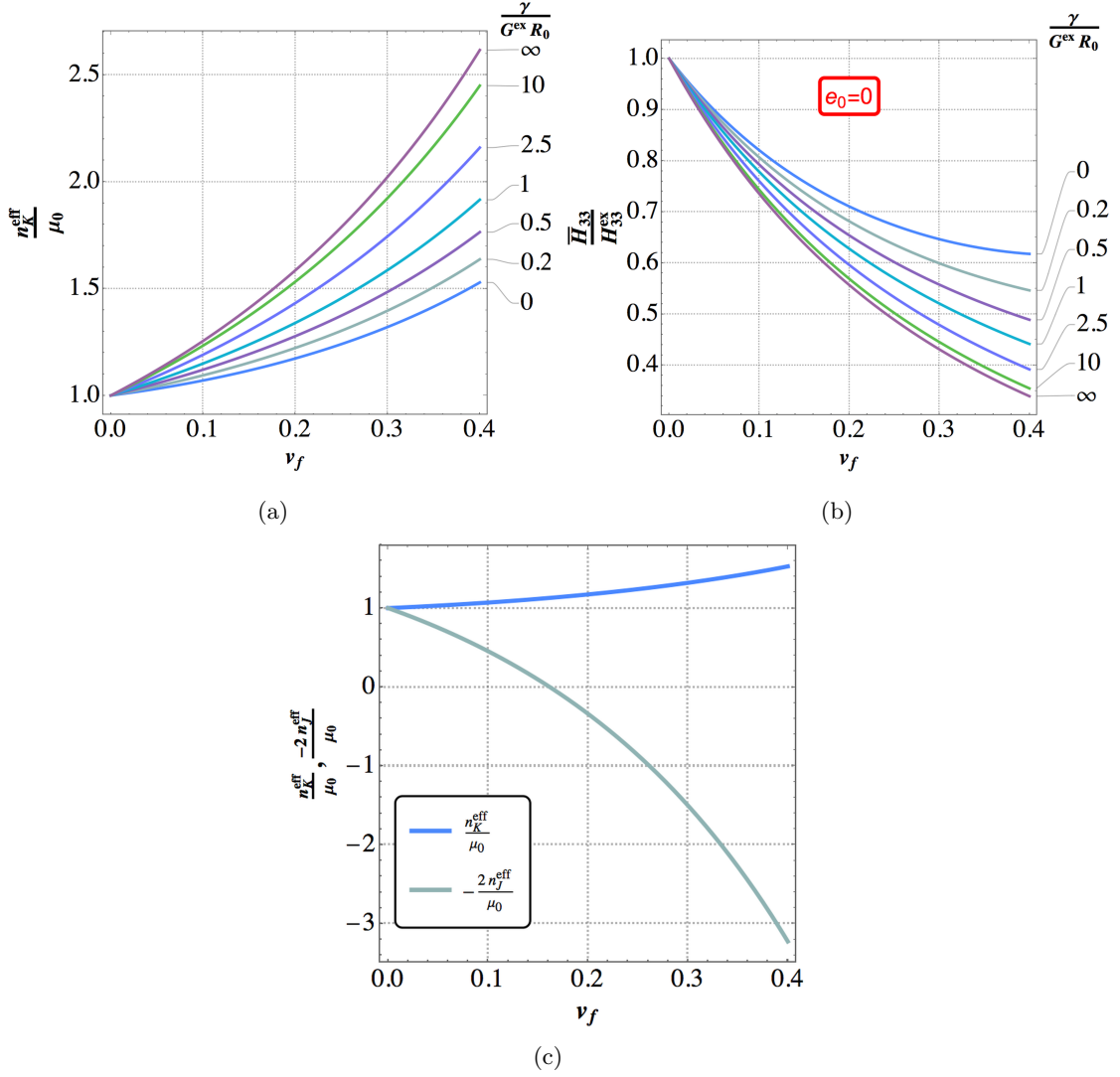


Figure 4.4.5: Effective magnetostrictive properties of an incompressible matrix filled with liquid-like magnetic spherical particles. ($\kappa^{\text{ex}} = +\infty, G^{\text{in}} = 0, \kappa^{\text{in}} = +\infty, \mu^{\text{in}} = 10\mu_0$)

to control electric polarization of the composite soft material merely by applying an external magnetic field. This is the heart of the magnetoelectric effect. We also note that increasing the surface tension of the liquid like inclusions results in a strong magnetoelectric response. This result is very promising. It is possible to create a soft material composite that shows magnetoelectric effect simply by adding soft liquid inclusions, with magnetic permeability different than μ_0 , to a soft dielectric matrix. If we choose inclusion size and composition carefully in order to allow larger surface tension at the matrix/inclusion interface, the mag-

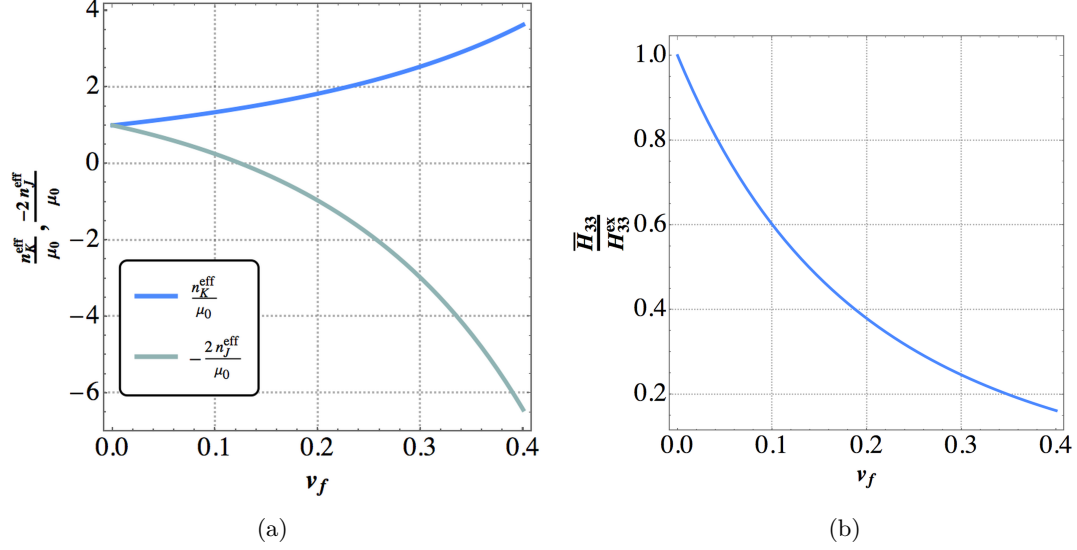


Figure 4.4.6: Effective magnetostrictive properties and deformation of an incompressible matrix filled with stiff magnetic spherical particles. $\kappa^{\text{ex}} = +\infty, G^{\text{in}} = +\infty, \kappa^{\text{in}} = +\infty, \mu^{\text{in}} = 100\mu_0$.

netoelectric response of our composite would be even stronger.

Next, we report the magnetostrictive properties of an incompressible matrix filled with liquid magnetic spherical particles. The materials properties used to represent our results are realistic for a typical dielectric elastomer and inclusion made of ferrofluid. In the absence of the electric field, we study the effect of the surface tension and the volume fraction of the inclusion on the magnetostrictive response. First, we note that magnetostrictive response increases as volume fraction of the inclusion increases. Adding more liquid inclusions to a soft matrix results in larger deformation upon the application of a magnetic field. Similarly, the increased surface tension makes the composite more magnetically responsive.

Moreover, we look at the opposite limit of stiff magnetic particles in Fig.4.4.6, such as inserting tiny iron particles inside a soft matrix. We notice in this case the effect of including surface tension is negligible. It does not affect the magnetostriction deformation of the overall composite.

Finally, we graph the deformation of the composite under the application of external electric and magnetic fields in order to understand their effect on the overall deformation as function of the volume fraction. We fix the magnitude of $\bar{\mathbf{e}}, \bar{\mathbf{h}}_0$ and we vary the surface

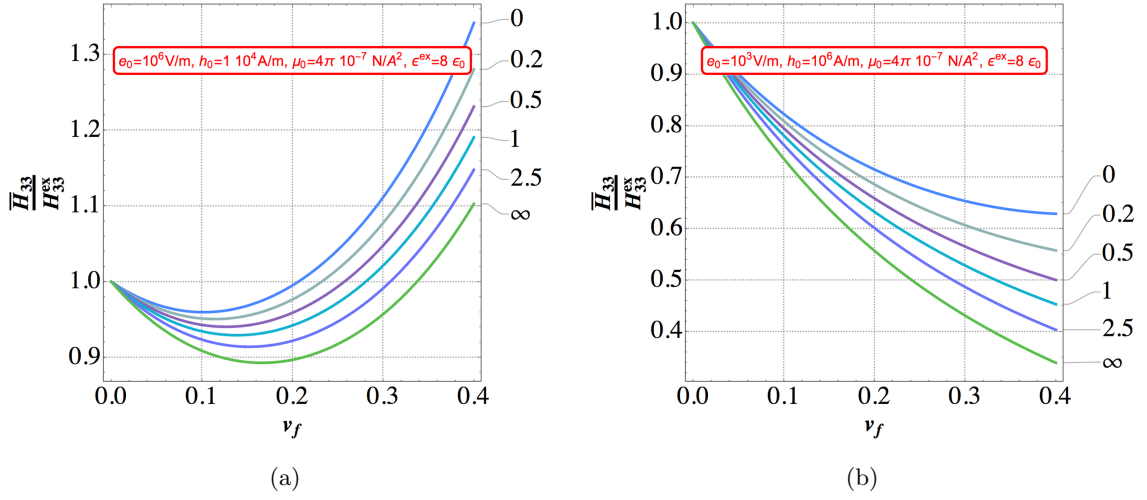


Figure 4.4.7: Effective magnetostrictive deformation of an incompressible matrix filled with liquid-like magnetic high permittivity spherical particles. $\kappa^{\text{ex}} = +\infty, G^{\text{in}} = 0, \kappa^{\text{in}} = +\infty, \mu^{\text{in}} = 10\mu_0, \epsilon^{\text{in}} = 100\epsilon^{\text{ex}}$

tension. In Fig.4.7(a), the magnitude of the electric field is high. We note for $v_f < 0.1$, the deformation is mainly dominated by the magnetic field since the normalized deformation decreases as surface tension γ increases. This matches the effect found earlier in Fig.4.5(b). However, when $v_f > 0.1$, the deformation increase as volume fraction increase for a constant surface tension. This have been reported in [78] for electrostriction deformation. For a weaker electric field and a stronger magnetic field as in Fig.4.7(b), the deformation is mainly dominated by the magnetic field. In short, there is a competition between the electric and magnetic field in controlling the overall deformation of the composite. Taking this into consideration, along with the effects of volume fraction and surface tension, it is possible to engineer a soft composite that responds in a specific way to external electric or magnetic field for technological applications.

4.5 Conclusion and Future Directions

In this chapter, we have presented a mathematical formulation to explore the behavior of a composite made with soft dielectric and liquid or solid spherical inclusion. The elastic contribution from surface tension was also taken into account. We report an enhanced

magneto-electro-elastic response that depends on volume fraction and surface tension particularly in the case of liquid inclusions. This setting was not reported yet in the experimental research work tackling inclusions. We have shown that the composite exhibit a strong apparent magnetoelectric behavior that can be used for technological applications. Our work provides basis to further study magnetoelectric properties for these composites experimentally in order to design soft "apparent" magnetoelectric materials.

Chapter 5

Microscopic Mechanisms Underpinning Flexoelectricity in Soft Materials

Flexoelectricity, the development of polarization upon changes in strain gradients, is a universal electromechanical coupling mechanism that appears in crystalline as well as soft materials. It has attracted immense attention since it allows the realization of several interesting applications. While the microscopic origins of flexoelectricity in hard crystalline materials are well-understood, a similar insight into soft material is currently missing. Inspired from the experimental and theoretical exploration of similar such mechanisms in the context of piezoelectricity in polymers, we propose that the presence of frozen dipoles and their thermal fluctuation is the root cause of flexoelectricity in soft materials. In our work, and following an Onsager cavity approach, we calculate the flexoelectric coefficient of soft electret structures as the change in the polarization of frozen dipoles as function of curvature. We found out that the result depends on the change of the volume with respect to curvature in addition to other known physical properties. We believe that this explains the wide range of flexoelectric coefficients experimentally reported which contradicted previous theoretical predictions. Moreover, we explore the temperature dependence

of the flexoelectric effect and compare it to experimental measurements.

5.1 Introduction

A macroscopic strain gradient can induce electric polarization in all materials. This phenomenon, known as *flexoelectricity*, is a universal and converse electromechanical coupling mechanism that is more general than piezoelectricity. The latter is the change in electric polarization due to stress. The piezoelectric effect has been extensively studied due to its promising applications that range from energy harvesters to self powered sensors [9]. However, the flexoelectric effect was understudied and started getting attention fairly recently. Because of its microscopic origins that require the material to be non-centrosymmetric, piezoelectric effect exists in certain materials only. On the other hand, the flexoelectric effect exists in all materials. Mathematically, the change in polarization due to an applied mechanical strain is given by

$$P_i \sim d_{ijk}\epsilon_{jk} + f_{ijkl}\frac{\partial\epsilon_{jk}}{\partial x_l}, \quad (5.1.1)$$

where P_i accounts for the component of the polarization vector, ϵ_{jk} is the component of the strain tensor, d_{ijk} refer to the components of the third order piezoelectric tensor and f_{ijkl} are the components of the fourth order flexoelectric tensor. In the case of a centrosymmetric crystal, the components of the piezoelectric tensor vanish. Since fourth order material property tensors are generally permissible regardless of the material symmetry, the flexoelectric tensor is non-zero for all materials. This mathematical formulation confirms that flexoelectricity is indeed universal. The flexoelectric effect was experimentally confirmed in several centrosymmetric crystals such as NaCl, strontium titanate, barium titanate among others [97, 98, 99, 100] which otherwise are not piezoelectric.

The flexoelectric effect initially appeared in liquid crystals. Later, extensive theoretical and experimental research work has been done on flexoelectricity in both crystalline and soft materials [10, 11, 97, 98, 99, 100, 101]. The close dependence of strain gradient on material size allows size dependent effects especially at the nanoscale. As a result, exploiting flexoelec-

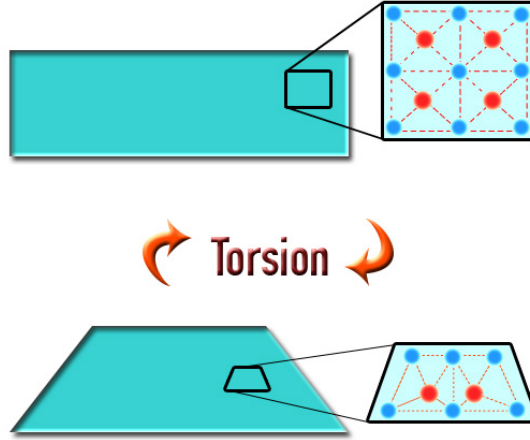


Figure 5.1.1: Microscopic origin of induced polarization in a crystalline material upon the application of a strain gradient

tricity will undoubtedly unravel several tantalizing applications such as creating artificial piezoelectric materials, giant and size dependent piezoelectricity in inhomogeneously deformed nanomaterials, energy harvesting, understanding hearing mechanism, ion transport in addition to others [9].

The microscopic origins of flexoelectricity in crystalline materials have been clearly understood. In the absence of any sources of external stress, the centers of positive and negative charges in a molecule coincide; thus making the total polarization null. The situation does not change in centrosymmetric crystals if a uniform strain is applied. However, if a non-uniform strain is applied, such as torsion or bending, the centers of positive and negative charges shift. Therefore, a dipole is created in the direction of the strain gradient resulting in an electric polarization as in Fig.5.1.1.

On the other hand, the mechanisms of flexoelectricity in polymers are still unclear. Taganstev suggested that the flexoelectric coefficient of crystalline materials is linearly proportional to the dielectric susceptibility of the material χ_{ij} and have an order of magnitude of e/a (e being the unit electronic charge and a the atomic dimension of the unit cell), i.e, $f_{ijkl} \sim \chi_{ij}(e/a)$ [102]. A similar relationship was suggested by Marvan for polymers as well

[103, 104]. Since the dielectric constant k of PVDF-based polymer materials, for instance, is small $k < 100$, and $e/a \sim 0.4nC/m$, the flexoelectric effect is expected to be very weak in soft materials. This dependence was initially confirmed experimentally [105]. Flexoelectric coefficients for polymers were found to be small $\sim 10^{-11} - 10^{-10}C/m$. However, some recent experimental investigations have reported a much larger order of magnitude, thus contradicting the previous results. The first giant flexoelectric effect was reported on bent-core liquid crystal elastomers with the nematic phase [106]. Moreover, latest measurements of the flexoelectric coefficients for PVDF [107, 108, 109, 110] revealed values much larger than anticipated $\sim 10^{-6}C/m$. Further studies looked at the flexoelectric coefficients of other polymers such as PET, polyethylene and epoxy [111, 112]. The results came out to be independent of the dielectric constants and in the range of $\sim 10^{-8} - 10^{-9}C/m$.

Based on these new findings, we suggest that the presence of frozen molecular dipoles and their thermal fluctuations, normally affected by experimental processing, is a main contributor to the flexoelectric response in soft materials. In this context, we calculate the change in the polarization of frozen dipoles in a soft electret structure as function of curvature following an Onsager cavity approach. We prove that the final result depends explicitly on the pre-existing dipole polarization and on the change of volume with respect to curvature in addition to other known physical properties. We also show that this contribution when added to the intrinsic flexoelectricity of the polymer explains the broad range of flexoelectric constants recently reported. In fact, we believe that preparation temperature and methods in addition to other experimental set-up conditions in the aforementioned studies may have significantly affected the magnitude of the flexoelectric coefficients for polymers. In addition, we explore the temperature dependence of the flexoelectric coefficient. We find out that flexoelectric behavior decreases as a function of temperature and we compare our results to some experimental values that have been already reported.

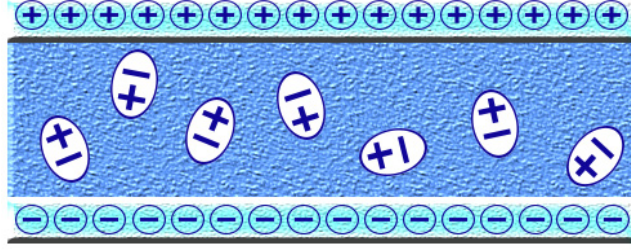


Figure 5.2.1: Randomly oriented molecular dipoles in an amorphous polymer

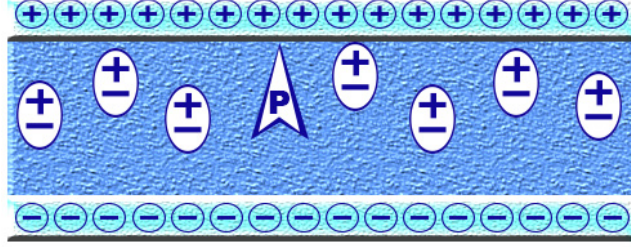


Figure 5.2.2: Molecular dipoles reoriented after the application of an electric field resulting in a permanent polarization

5.2 Flexoelectric Coefficient For Molecular Dipole Electrets.

We start by considering an amorphous polymer containing molecular dipoles that have been partially aligned under the application of an external electric field. Cooling the sample below the glass transition temperature makes those dipoles frozen and results in a permanent polarization. Here, we follow a statistical mechanics approach and cavity reaction field calculation that was introduced by Onsager. Initially, we assume that the polymer contains randomly oriented molecular dipoles with permanent moment μ and polarizability α . The electric permittivity of the polymer is taken to be ϵ .

After cooling, the average polarization is no longer zero. The resultant permanent polarization P is given by [113]

$$P = \frac{(\epsilon + 2)}{3} \frac{N}{V} \mu_0 \langle \cos \gamma \rangle, \quad (5.2.1)$$

where ϵ_L accounts for the liquid dielectric constant, N is Avogadro's number, V is the volume, μ_0 is the magnitude of the permanent dipole moment and γ is the angle between

the direction of the permanent polarization and the individual dipole axis. With equation (5.2.1) it is possible to calculate the change in total polarization due to the change in any external parameter X such as volume, temperature, pressure or curvature as follows:

$$\begin{aligned} \frac{\partial P}{\partial X} = & -\frac{N}{V_0^2} \frac{\epsilon + 2}{3} \mu_0 \langle \cos \gamma \rangle \frac{\partial V}{\partial X} + \\ & \frac{N}{3V_0} \mu_0 \langle \cos \gamma \rangle \frac{\partial \epsilon}{\partial X} + \frac{N}{V_0} \frac{\epsilon + 2}{3} \mu_0 \frac{\partial \langle \cos \gamma \rangle}{\partial X}. \end{aligned} \quad (5.2.2)$$

Before we proceed, we can express the second term as a derivative of the volume V , rather than ϵ , using the following relationship between dielectric permittivity and volume [114]:

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{C}{V} - \frac{D}{V^2}, \quad (5.2.3)$$

where C and D are constants for the material and can be found from experimental measurements. Carrying out the differentiation with respect to any parameter X , we get

$$\frac{\partial \epsilon}{\partial X} = -\frac{1}{V_0} \left(\frac{(\epsilon + 2)(\epsilon - 1)}{3} - \frac{D}{V_0^2} \frac{(\epsilon + 2)^2}{3} \right) \frac{\partial V}{\partial X}. \quad (5.2.4)$$

D in the above equation can be estimated experimentally and is relatively small.

Substituting eq. (5.2.4), we can rewrite eq. (5.2.2) as

$$\begin{aligned} \frac{\partial P}{\partial X} = & -\frac{N}{V_0^2} \mu_0 \langle \cos \gamma \rangle \frac{\partial V}{\partial X} \left(\frac{\epsilon + 2}{3} + \frac{(\epsilon + 2)(\epsilon - 1)}{9} \right. \\ & \left. - \frac{D}{V_0^2} \frac{(\epsilon + 2)^2}{3} \right) + \frac{N}{V_0} \frac{\epsilon + 2}{3} \mu_0 \frac{\partial \langle \cos \gamma \rangle}{\partial X}. \end{aligned} \quad (5.2.5)$$

When a strain gradient is applied to the polymer sample, we expect an induced electric polarization that would result in flexoelectric effect. In this work, we assume that the polymer sample is undergoing bending conditions. As such, we replace X in eq. (5.2.5) by curvature κ and calculate the flexoelectric coefficient f as:

$$f = \frac{\partial P}{\partial \kappa}. \quad (5.2.6)$$

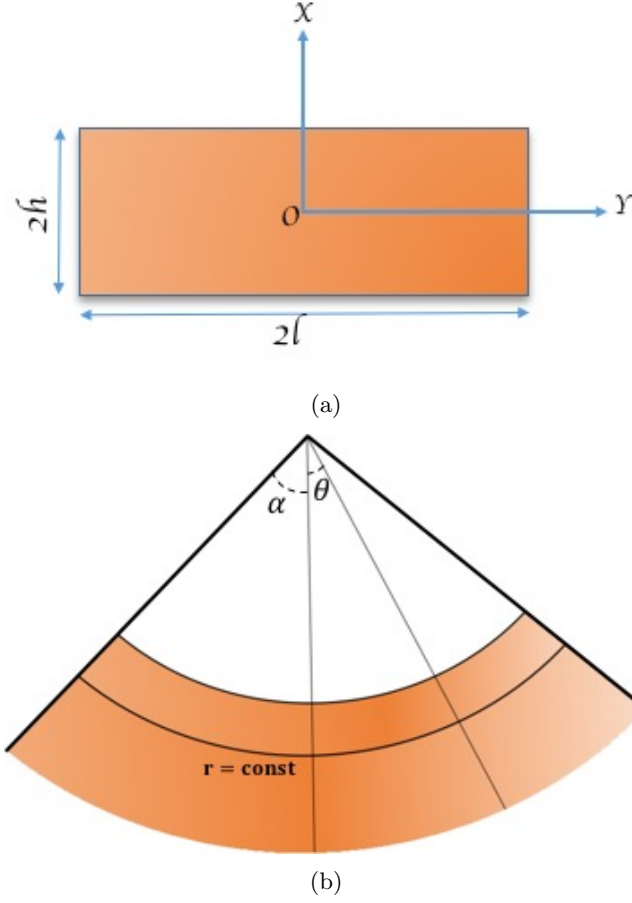


Figure 5.2.3: (a) Rectangular block in an underformed state, (b) Bent rectangular block in the deformed state

∂P is the induced polarization that results from bending.

In order to calculate ∂P , we consider two states. The first one is the flat polymer with pre-existing polarization P_0 that can be found from eq. (5.2.1). The second state is a bent polymer with curvature κ . Our purpose is to find the change in polarization that results between the two configurations to be able to estimate the flexoelectric response due to bending. As such, we need to evaluate the change of the volume V and angle γ with respect to curvature κ .

5.2.1 Change of volume with respect to curvature

Upon applying mechanical strain, the volume of any body in the deformed configuration is related to its volume in the reference configuration through the volumetric ratio J through

$$V = JV_0, \quad (5.2.7)$$

where V_0 is the volume in the undeformed state. In this setting, we are interested in the finite bending of a rectangular block. The purpose is to find J so we can evaluate the change in the volume of the block after bending.

In the reference configuration, Fig.5.3(a), we use a cartesian coordinates system $OXYZ$ with basis $\mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3$ in the direction of OX, OY and OZ respectively. The rectangular block is denoted by

$$\Omega = \{(X, Y, Z) \in \mathbb{R}^3 : |X| \leq h, \quad |Y| \leq l, \quad |Z| \leq t\}. \quad (5.2.8)$$

In the deformed configuration, we assume that the rectangular block is bent into a sector of a cylinder as in Fig.5.3(b). We use cylindrical coordinates $or\theta z$ to describe the deformation such that a planes $X = \text{const}$ and $Y = \text{const}$ in the reference configuration become the planes $r = \text{const}$ and $\theta = \text{const}$ respectively in the deformed configuration. The basis in cylindrical polar coordinates is defined as $\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_z$ where $\mathbf{e}_z = \mathbf{E}_3$. Since the deformation is relatively large and cannot be considered infinitesimal, we will use Hencky's model to account for a general compressible deformation. Generally, to describe the deformation of the block, we use

$$r = r(X), \quad \theta = \frac{\alpha}{l}Y, \quad z = \lambda z, \quad (5.2.9)$$

where 2α is the bending angle and λ is the stretch normal to the bending plane. The radial and the circumferential stretches are denoted by λ_r and λ_θ respectively. They are defined as

$$\lambda_r = \frac{dr}{dX}, \quad \lambda_\theta = \frac{\alpha}{l}r. \quad (5.2.10)$$

The deformation gradient $\mathbf{F}$ is given by

$$\mathbf{F} = \lambda_r \mathbf{e}_r \otimes \mathbf{E}_1 + \lambda_\theta \mathbf{e}_\theta \otimes \mathbf{E}_2 + \lambda_{\mathbf{e}_z} \otimes \mathbf{E}_3. \quad (5.2.11)$$

Thus, the volumetric ratio J is

$$J = \det \mathbf{F}. \quad (5.2.12)$$

This problem has been solved in reference [115, 116]. Following the calculations therein, J is given by

$$\begin{aligned} J &= (\lambda \lambda_\theta)^{(1-2\nu)/(1-\nu)} \exp(1 - \sqrt{\Phi}) \\ \Phi &= 1 + \frac{1-2\nu}{(1-\nu)^2} \left((\ln \lambda_\theta^- + \nu \ln \lambda)^2 - (\ln \lambda_\theta + \nu \ln \lambda)^2 \right), \end{aligned} \quad (5.2.13)$$

where ν is the Poisson ratio of the material and λ_θ^- is the minimum circumferential stretch at $r = r|_{X=-h}$. If $\nu = 0.5$, in eq. (5.2.13), $J = 1$ and that corresponds to the incompressible deformation case. For any ν that is different than 0.5, the deformation is compressible. The dependence of the volumetric ratio J on curvature κ appears through λ_θ . Taking the average curvature of the cylindrical sector to be the $\kappa = \frac{1}{R}$ where R is the radius of the middle plane, we can rewrite the eq. (5.2.10)₂ for the middle plane as

$$\lambda_\theta = \frac{\alpha}{l} R = \frac{\alpha}{l} \frac{1}{\kappa}. \quad (5.2.14)$$

Thus, the change of volume resulting from bending is

$$\frac{\partial V}{\partial \kappa} = \frac{\partial J}{\partial \kappa} V_0, \quad (5.2.15)$$

where J is given by eq. (5.2.13).

5.2.2 Change of the dipole orientation angle with respect to curvature

Next, we are interested in exploring the dependence of $\langle \cos \gamma \rangle$ on the curvature κ . For the undeformed configuration, the molecular dipoles are assumed to behave like classical rotational harmonic oscillators [113] where γ is the angle of oscillation with respect to the

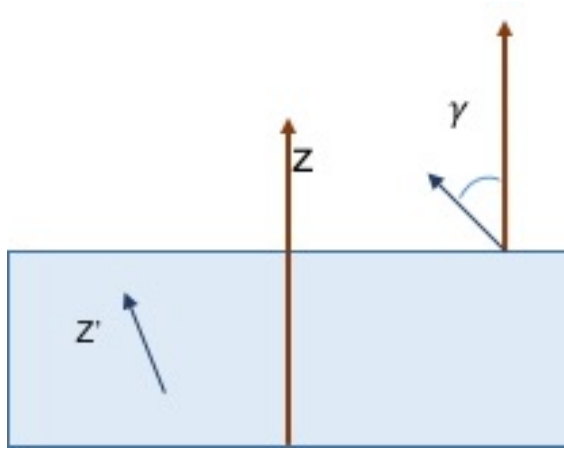


Figure 5.2.4: Angle γ is defined as the angle between the permanent polarization direction z and z' , the average position of one dipole

permanent polarization direction (Fig.5.2.4). Assuming z to be the polarization direction, z' is the direction of the average position of one dipole, then $\langle \cos \gamma \rangle$ would represent the projection of the unit vector representing the dipole along the z axis. If we choose the x' axis so the dipole is oscillating with amplitude ϕ_0 about the y' axis, then the dipole remains in the $x'z'$ plane. Using the definition of the Eulerian angles and averaging, we get the following expression for $\langle \cos \gamma \rangle$ [113]:

$$\langle \cos \gamma \rangle = \cos(\gamma) J_0(\phi_0), \quad (5.2.16)$$

where $J_0(\phi_0)$ is the Bessel function of first kind and order zero.

Upon bending the dielectric film, the angle γ_0 between the average position of one dipole and the polarization direction in the flat state changes. The new angle γ depends on the old angle γ_0 and the bending angle α . Thus, we will have

$$\begin{aligned} \langle \cos \gamma \rangle &= \cos(\gamma) J_0(\phi_0) \\ &= \cos(\gamma_0 + \alpha) J_0(\phi_0) \\ &= [\cos(\gamma_0) \cos(\alpha) - \sin(\gamma_0) \sin(\alpha)] J_0(\phi_0). \end{aligned} \quad (5.2.17)$$

If we assume that horizontal component of the dipole polarization averages to zero, and using eq.(5.2.16) then we are left with

$$\langle \cos \gamma \rangle = \cos(\gamma_0) \cos(\alpha) J_0(\phi_0). \quad (5.2.18)$$

Carrying out the differentiation with respect to curvature κ , we get

$$\begin{aligned} \frac{\partial \langle \cos \gamma \rangle}{\partial \kappa} = & -\cos(\gamma_0) \cos(\alpha) J_1(\phi_0) \frac{\partial \phi_0}{\partial \kappa} \\ & - \cos(\gamma_0) J_0(\phi_0) \sin(\alpha) \frac{\partial \alpha}{\partial \kappa}. \end{aligned} \quad (5.2.19)$$

In order to find the first derivative in eq.(5.2.19), we use the following expression for the amplitude of a classic rotational harmonic oscillator:

$$\phi_0^2 = \frac{2k_b T}{I\omega^2}, \quad (5.2.20)$$

where k_b is the Boltzmann constant, T is the temperature, I is the moment of inertia and ω is the frequency of vibration. The latter depends solely on volume though the Grüneisen constant $\gamma_g = -(V/\omega)(d\omega/dV)$. Thus, assuming constant temperature, we get:

$$\frac{\partial \phi_0}{\partial \kappa} = \frac{\phi_0}{V_0} \gamma_g \frac{\partial V}{\partial \kappa}. \quad (5.2.21)$$

As for the second derivative in eq. (5.2.19), we can re-arrange eq. (5.2.14) $\alpha = \lambda_\theta \frac{l}{R} = \lambda_\theta l \kappa$ to find it. Finally, using eq. (5.2.1), (5.2.13), (5.2.15), (5.2.19), and (5.2.21), eq.(5.2.5) can be written as

$$\begin{aligned} \frac{\partial P}{\partial \kappa} = & -P_0 \frac{\partial J}{\partial \kappa} \left[1 + \frac{\epsilon - 1}{3} - \frac{D}{V_0^2} (\epsilon + 2) \right. \\ & \left. + \cos \alpha \phi_0 \gamma_g \frac{J_1(\phi_0)}{J_0(\phi_0)} \right] - P_0 \sin \alpha \frac{\partial \alpha}{\partial \kappa}. \end{aligned} \quad (5.2.22)$$

If we take a close look at eq. (5.2.22), we notice that the final expression depends on three components. The first one arises from the change of volume with respect to curvature. The second one comes from the deformation and depends on the bending angle whereas the last effect stems from the pre-existing polarization. Moreover, we remark that there exists an implicit temperature dependence that emerges from the permittivity of the dielectric

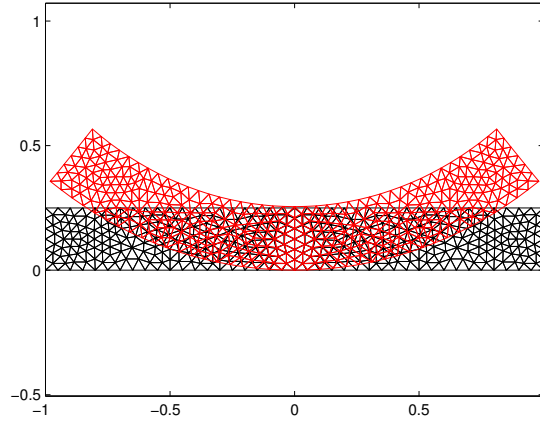


Figure 5.3.1: FEM simulation of the deformed beam for $\kappa = 0.1$ using numerical procedure describe earlier

material ϵ and the amplitude of vibration ϕ_0 .

5.3 Results and Discussion

To get a better understanding of the change in polarization upon bending, we employ the numerical method suggested recently by reference [117]. A domain $\Omega \in [-L/2; L/2] \times [-H/2; H/2]$ is meshed with quadratic six-node triangular finite elements as in Fig.5.3.1 . We choose $\lambda = 1$ (plane strains), $L = 1$, $H = 1/4$. The external nodes of the domain are submitted to boundary conditions that prescribe the exact solution of a hyperelastic Hencky model subjected to bending [116]. The preexisting polarization is $P_y^0 = 10^{-4}$ C/m². To prescribe bending, all nodes of the boundary of the domain are subjected to $\mathbf{u} = (\bar{\mathbf{F}} - \mathbf{1}) \mathbf{x}$, such that $\mathbf{x} \in \Omega$ and $\bar{\mathbf{F}} = \mathbf{R}\mathbf{U}$, where

$$\mathbf{U} = \begin{bmatrix} \lambda_\theta & 0 \\ 0 & \lambda_r \end{bmatrix}, \quad \mathbf{R} = \begin{bmatrix} \cos(\theta) & -\sin(\theta) \\ \sin(\theta) & \cos(\theta) \end{bmatrix}, \quad (5.3.1)$$

where θ is defined in eq.(5.2.9) and $\lambda_\theta, \lambda_r$ defined in eq.(5.2.10).

The polarization P_y in the deformed beam is plotted in Fig.5.3.2. It is clear from this figure that distribution of polarization changes after bending.

We graph the effect of change in curvature on the volume of the block in Fig.5.3.3 using

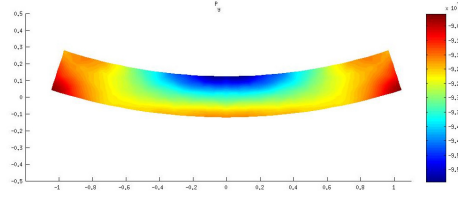


Figure 5.3.2: Polarization in the deformed beam

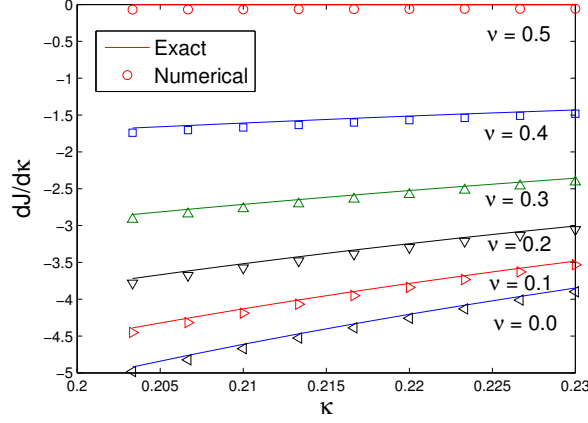


Figure 5.3.3: Variation of volume versus curvature κ for different Poisson's ratios

eq.(5.2.15). We expect this term to have a crucial effect if the material is compressible, i.e. $\nu \neq 0.5$. We validate our results using the numerical procedure described above.

We note that for incompressible deformation $\nu = 0.5$ the change in volume due to the deformation is zero, which is expected. In case of incompressibility, there is no contribution coming from the change in the volume. For Poisson's ratio different than 0.5, the change in volume depends significantly on the curvature. We also note that the slope of the curve becomes steeper as the Poisson's ratio decreases. This is a very prominent conclusion particularly for the case of a highly incompressible material like porous polymers, the change in volume upon bending has a major contribution to the induced polarization.

Additionally, it is clear that the final expression (eq.(5.2.22)) depends explicitly on the polarization of the preexisting dipoles in addition to other material parameters. This proves our initial speculation that presence of molecular dipoles and their thermal fluctuation could be a major factor behind widely varying experimental reports of flexoelectric constants for several polymer samples. In fact, the total flexoelectric response of a polymer is the sum of

an intrinsic component and another component related to the presence of molecular dipoles and the resulting permanent polarization. Thus, we can find the flexoelectric coefficient of a polymer using

$$f = f^{int} + f^{dipoles}. \quad (5.3.2)$$

The first contribution on the right hand side corresponds to the intrinsic response that was predicted to depend on the dielectric susceptibility and charge to atomic dimension ratio e/a [102, 103, 104]. This is typically small in magnitude for soft materials. The other term on the right hand side corresponds to what has been calculated above in eq.(5.2.22). Experimental preparation of polymer samples and processing temperatures will significantly influence the magnitude of this term. This can be confirmed merely by looking at sample characterization coming from different sources. Unexpectedly giant flexoelectric constants could be due to the presence of a great number of partially oriented molecular dipoles that create a large permanent polarization, thus enhancing the flexoelectric response of the polymer sample. On the other hand, a small flexoelectric constant results from the absence of these dipoles or their random orientation inside the sample which significantly reduces their overall influence on the behavior of the material in the presence of a strain gradient.

We show some sample calculations to elucidate the effect of preexisting polarization on the flexoelectric response. We assume a Poisson's ratio $\nu = 0.35$, dielectric constant $\epsilon = 9$, $\phi_0 = 10^\circ$, and a curvature of 0.2. If the polarization of pre-existing dipoles is around $10^{-6}C/m^2$, the flexoelectric coefficient as calculated from our model would be on the order of $10^{-10}C/m$. This order of magnitude matches the order measured for PVDF by Poddar and Ducharme [108] and for some elastomers by Marvan [105]. For a pre-existing polarization on the order of 10^{-4} , the flexoelectric coefficient is higher and on the order of $10^{-8}C/m$. Experiments by Chu and Salem [112] on several thermoplastic and thermosetting polymers have reported the similar order of magnitude. Higher values of polarization would yield a larger order of magnitude.

The purpose here is to provide an intuition on the reason behind the wide range of experimental values reported on flexoelectricity. For instance, PVDF is a highly dipolar ferroelectric [108]. In fact, an experimental study on the variation of polarization of PVDF

as function of the different temperature and external field treatments revealed that electromechanical coupling constant can be greatly enhanced by the set-up conditions [118]. Moreover, PVDF comes in various phases such as α or β phases which changes the crystallinity percentage. It can also have different Poisson's ratios depending on porosity. All of these conditions, which appear in our final result eq.(5.2.22), contribute in the flexoelectric behavior of the used sample.

It is actually possible to increase the pre-existing polarization by poling a polymer. When a polymer undergoes electric poling, the resulting polarization depends on the poling field E_p through the relation:

$$P_0 = \delta\epsilon\epsilon_0 E_p. \quad (5.3.3)$$

A strong poling field results in more molecular dipoles aligned with the direction of the applied electric field, and thus a stronger permanent polarization, whereas a small poling field results in a smaller permanent polarization.

Finally, we explore the change in flexoelectric response as function of the temperature. Looking at eq. (5.2.22), the temperature dependence of the flexoelectric coefficient is not explicit. However, if we examine the terms in that equation, we note that the electric permittivity ϵ and the amplitude of vibration of dipole ϕ_0 are temperature dependent. We can find experimental research work that explored the change of ϵ as a function of temperature in the literature (see for example [119]). The dependence of ϕ_0 on temperature arises from statistical mechanics eq.(5.2.20). Moreover, the Poisson ratio is also temperature dependent, which in turn affects the compressibility of the material, i.e. eq.(5.2.15).

We use data provided from experiments in order to get a tentative temperature dependence of flexoelectricity. We plot the change of the flexoelectric coefficient as function of temperature for P(VDF-TrFE-CFE) terpolymer and compare it to the experimental values obtained in reference [109]. The pre-existing polarization P_0 in eq.(5.2.22) is taken as 1.8×10^{-3} as in the reference, the curvature is chosen such that the strain gradient is $1m^{-1}$, the values for Poisson's ratio as function of temperature are obtained from reference [119], $\phi_0 = 10^\circ$ [113] and the values of the dielectric constant as function of temperature are obtained from reference [109]. The temperature dependent flexoelectricity as calculated from eq.(5.2.22) is

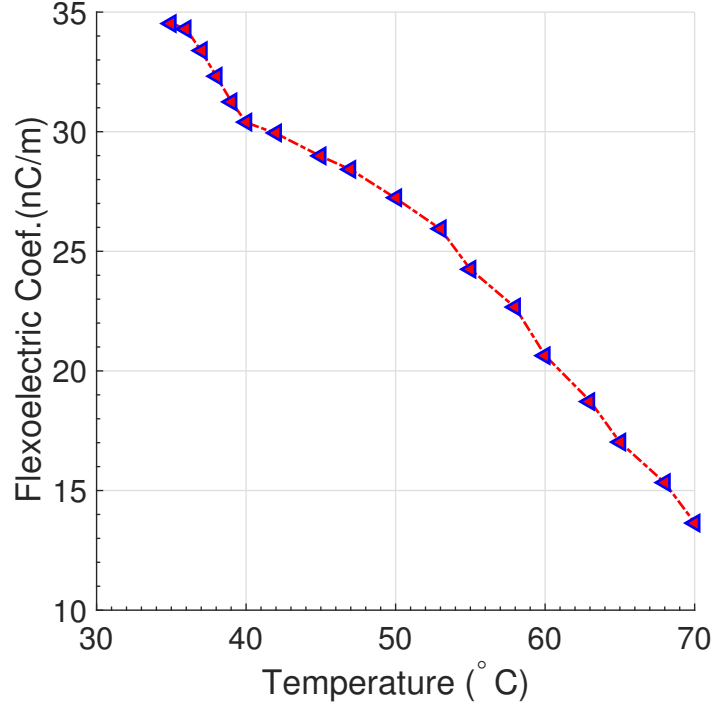


Figure 5.3.4: Temperature dependence of flexoelectricity

given in Fig.5.3.4. We note that our model predicts a decrease in the flexoelectric coupling as function of temperature. This is confirmed by experimental measurements. As a matter of fact, the flexoelectric coefficient depends proportionally on the dielectric constant which decreases as temperature increases in this case. Comparing the trend of our graph to experimental data, we find that they have different behavior for temperature below $\sim 45^\circ\text{C}$. In our work, we are strictly computing the change in polarization due to the flexoelectric effect without any piezoelectric contribution. Our model does not show any dependence on the elastic modulus. As a result, we cannot capture the increasing trend reported in experiments.

5.4 Conclusion

In summary, we have provided an insight on the wide range of flexoelectric coefficients measured in soft materials. Following an Onsager cavity approach, we derived the flexoelectric coefficient of an amorphous polymer as the change in preexisting polarization with

respect to the change in curvature. Our final result shows explicit dependence on the total polarization of frozen dipoles, the change of volume due to deformation in addition to other materials parameters. This confirms that the microscopic mechanism of flexoelectricity in polymers is indeed more complex than it was predicted. The flexoelectric response of a soft dielectric material does not simply depend on the dielectric constant, but is vulnerable to several conditions. With this understanding, giant flexoelectric behavior in soft polymers can be engineered by selecting the appropriate parameters and following experimental procedures to incorporate frozen molecular dipoles during the fabrication process.

Chapter 6

Concluding Remarks

In this dissertation, we have addressed several issues related to electro-magneto-mechanical coupling in soft dielectric materials. The magnetoelectric effect in soft one phase dielectric materials and composites was the main focus in chapters 2, 3 and 4. In chapter 5, we discussed a very interesting electromechanical coupling in soft materials which is flexoelectricity. Specifically, the following results are obtained in these contexts:

- We investigated the stability of a dielectric material subject to a mechanical strain, an electric voltage and a magnetic field. We highlighted the effect of the magnetic field on enhancing the stability of dielectric material even in the simple case of a thin film and homogeneous deformation. As a result of the external magnetic field, the dielectric film can sustain larger electric field before instability kicks in. We have used this effect to suggest a simple idea for wireless energy harvesting where it is possible to generate electric power using stray magnetic fields.
- We proposed a relatively simple concept to engineer soft materials that shows concurrent apparent piezoelectricity and magnetoelectricity using electret structure. Geometric non-linearity and the presence of stable charges at the interfaces of the material is the key mechanism to couple mechanical, electric and magnetic fields. We have also illustrated this result using some known polymer materials that have experimentally shown stable electret structure.

- We explored the effective mechanical, electric and magnetic properties of a composite material made with a soft polymer matrix and spherical inclusions that have high electric permittivity and magnetic permeability at the same time while considering capillary effect. We found that the composite reveals a magnetoelectric coupling effect that could be enhanced due to surface tension of the liquid inclusions. The results were particularly significant since it offers many tantalizing applications that require soft material with a magnetoelectric behavior such as soft robots, wireless energy harvesters, sensors and actuators in addition to others.
- We highlighted the microscopic mechanisms for the flexoelectric response in soft dielectric materials. We were able to explain the wide range of flexoelectric coefficients that have been reported experimentally. We showed that existence of molecular dipoles and their thermal fluctuations in addition to material properties are the root cause behind large and small flexoelectric coupling coefficients. We also predicted the effect of temperature on flexoelectricity. Taking those parameters into consideration allows the freedom to control the flexoelectric behavior of dielectric materials desired for specific applications.

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Appendix

We list the detailed derivations of the first variation (2.2.30) and the second variation (2.2.31) in the following.

Details of the first variation

Consider a smooth variation:

$$\delta \mathbf{v} = (\delta \lambda_2, \delta \lambda_3, \delta P, \delta M)^T \quad (6.0.1)$$

of the four generalized coordinates $\mathbf{v}$ in (2.2.23). Then the variation $\delta W^t(\mathbf{v})$ in (2.2.30) reads

$$\begin{aligned} \delta W^t(\mathbf{v}) &= \delta W(\mathbf{v}) - \epsilon_0(\lambda_2 \lambda_3 \xi_{,X}) \delta(\lambda_2 \lambda_3 \xi_{,X}) + \delta(\lambda_2 \lambda_3 \xi_{,X} P) \\ &\quad + \mu_0(\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}}) \delta(\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}}) - \mu_0 h^e \delta M, \end{aligned} \quad (6.0.2)$$

where

$$\delta W(\mathbf{v}) = \frac{\partial W}{\partial \lambda_2} \delta \lambda_2 + \frac{\partial W}{\partial \lambda_3} \delta \lambda_3 + \frac{\partial W}{\partial P} \delta P + \frac{\partial W}{\partial M} \delta M, \quad (6.0.3a)$$

$$\delta(\lambda_2 \lambda_3 \xi_{,X}) = (\lambda_3 \xi_{,X}) \delta \lambda_2 + (\lambda_2 \xi_{,X}) \delta \lambda_3 + (\lambda_2 \lambda_3) \delta \xi_{,X}, \quad (6.0.3b)$$

$$\begin{aligned} \delta(\lambda_2 \lambda_3 \xi_{,X} P) &= (\lambda_3 \xi_{,X} P) \delta \lambda_2 + (\lambda_2 \xi_{,X} P) \delta \lambda_3 + (\lambda_2 \lambda_3 P) \delta \xi_{,X} \\ &\quad + (\lambda_2 \lambda_3 \xi_{,X}) \delta P, \text{ and} \end{aligned} \quad (6.0.3c)$$

$$\delta(\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}}) = (\lambda_3 \zeta_{,X}^{\text{self}}) \delta \lambda_2 + (\lambda_2 \zeta_{,X}^{\text{self}}) \delta \lambda_3 + (\lambda_2 \lambda_3) \delta \zeta_{,X}^{\text{self}}. \quad (6.0.3d)$$

The terms related to the variation $\delta \xi_{,X}$ in (6.0.3b) and (6.0.3c) can be finally omitted by considering the Maxwell equation (2.2.21) and the variation of the electric boundary conditions as well as integration by parts.

Similarly, the term related to the variation $\zeta_{,X}^{\text{self}}$ in (6.0.3d) can also be recast, that is,

$$\begin{aligned}
& \int_0^{L_1} \mu_0(\lambda_2\lambda_3\zeta_{,X}^{\text{self}})(\lambda_2\lambda_3)\delta\zeta_{,X}^{\text{self}} dX \\
&= \int_0^{L_1} \left\{ \mu_0(\lambda_2\lambda_3\zeta_{,X}^{\text{self}})[(\lambda_2\lambda_3)\delta\zeta_{,X}^{\text{self}} - \delta M] + \mu_0(\lambda_2\lambda_3\zeta_{,X}^{\text{self}})\delta M \right\} dX \\
&= \int_0^{L_1} \mu_0(\lambda_2\lambda_3\zeta_{,X}^{\text{self}})\delta M dX.
\end{aligned} \tag{6.0.4}$$

The first term on the second line disappears due to the zero variation of the Maxwell equations (2.2.21) and the magnetic boundary condition with respect to the magnetization M .

Thus the first variation (2.2.30) can be written as

$$\frac{\delta F}{L_1 L_2 L_3} = \frac{1}{L_1} \int_0^{L_1} \left(\frac{\partial W^t}{\partial \mathbf{v}} \cdot \delta \mathbf{v} \right) dX - s_2 \delta \lambda_2 - s_3 \delta \lambda_3 = 0. \tag{6.0.5}$$

Here the vector derivative has the component form

$$\frac{\partial W^t}{\partial \mathbf{v}} = \left(\frac{\partial W^t}{\partial \lambda_2}, \frac{\partial W^t}{\partial \lambda_3}, \frac{\partial W^t}{\partial P}, \frac{\partial W^t}{\partial M} \right)^T, \tag{6.0.6}$$

where the components are

$$\frac{\partial W^t}{\partial \lambda_2} = \frac{\partial W}{\partial \lambda_2} - \epsilon_0 \lambda_2 \lambda_3^2 (\xi_{,X})^2 + \lambda_3 \xi_{,X} P + \mu_0 \lambda_2 \lambda_3^2 (\zeta_{,X}^{\text{self}})^2, \tag{6.0.7a}$$

$$\frac{\partial W^t}{\partial \lambda_3} = \frac{\partial W}{\partial \lambda_3} - \epsilon_0 \lambda_2^2 \lambda_3 (\xi_{,X})^2 + \lambda_2 \xi_{,X} P + \mu_0 \lambda_2^2 \lambda_3 (\zeta_{,X}^{\text{self}})^2, \tag{6.0.7b}$$

$$\frac{\partial W^t}{\partial P} = \frac{\partial W}{\partial P} + \lambda_2 \lambda_3 \xi_{,X}, \text{ and} \tag{6.0.7c}$$

$$\frac{\partial W^t}{\partial M} = \frac{\partial W}{\partial M} + \mu_0 (\lambda_2 \lambda_3)^2 \zeta_{,X}^{\text{self}} [\zeta_{,X}^{\text{self}}]_{,M} - \mu_0 h^e, \tag{6.0.7d}$$

$[\zeta_{,X}^{\text{self}}]_{,M}$ in (6.0.7d) is a coefficient related to the variations of the magnetization M and the self-magnetic field $\zeta_{,X}^{\text{self}}$ in (6.0.4).

Detailed second variation

Consider the integrand $\delta^2 W^t(\mathbf{v})$ in (2.2.31).

$$\begin{aligned}\delta^2 W^t(\mathbf{v}) &= \delta^2 W(\mathbf{v}) - \frac{\epsilon_0}{2} \delta^2 \left[(\lambda_2 \lambda_3 \xi_{,X})^2 \right] + \delta^2 [\lambda_2 \lambda_3 \xi_{,X} P] \\ &\quad + \frac{\mu_0}{2} \delta^2 \left[(\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}})^2 \right],\end{aligned}\tag{6.0.8}$$

where

$$\delta^2 W(\mathbf{v}) = \delta \mathbf{v} \cdot \mathbb{H}^1 \delta \mathbf{v},\tag{6.0.9a}$$

$$\begin{aligned}\delta^2 \left[(\lambda_2 \lambda_3 \xi_{,X})^2 \right] &= 2(\lambda_3 \xi_{,X})^2 \delta \lambda_2^2 + 2(\lambda_2 \xi_{,X})^2 \delta \lambda_3^2 \\ &\quad + 2(\lambda_2 \lambda_3)^2 \delta \xi_{,X}^2 + 8\lambda_2 \lambda_3 (\xi_{,X})^2 \delta \lambda_2 \delta \lambda_3 \\ &\quad + 8\lambda_2 \lambda_3^2 \xi_{,X} \delta \lambda_2 \delta \xi_{,X} + 8\lambda_2^2 \lambda_3 \xi_{,X} \delta \lambda_3 \delta \xi_{,X},\end{aligned}\tag{6.0.9b}$$

$$\begin{aligned}\delta^2 [\lambda_2 \lambda_3 \xi_{,X} P] &= 2 \left\{ \xi_{,X} P \delta \lambda_2 \delta \lambda_3 + \lambda_3 P \delta \lambda_2 \delta \xi_{,X} \right. \\ &\quad + \lambda_3 \xi_{,X} \delta \lambda_2 \delta P + \lambda_2 P \delta \lambda_3 \delta \xi_{,X} \\ &\quad \left. + \lambda_2 \xi_{,X} \delta \lambda_3 \delta P + \lambda_2 \lambda_3 \delta \xi_{,X} \delta P \right\},\end{aligned}\tag{6.0.9c}$$

$$\begin{aligned}\delta^2 \left[(\lambda_2 \lambda_3 \zeta_{,X}^{\text{self}})^2 \right] &= 2(\lambda_3 \zeta_{,X}^{\text{self}})^2 \delta \lambda_2^2 + 2(\lambda_2 \zeta_{,X}^{\text{self}})^2 \delta \lambda_3^2 \\ &\quad + 2(\lambda_2 \lambda_3)^2 \delta (\zeta_{,X}^{\text{self}})^2 + 8\lambda_2 \lambda_3 (\zeta_{,X}^{\text{self}})^2 \delta \lambda_2 \delta \lambda_3 \\ &\quad + 8\lambda_2 \lambda_3^2 \zeta_{,X}^{\text{self}} \delta \lambda_2 \delta \zeta_{,X}^{\text{self}} + 8\lambda_2^2 \lambda_3 \zeta_{,X}^{\text{self}} \delta \lambda_3 \delta \zeta_{,X}^{\text{self}}.\end{aligned}\tag{6.0.9d}$$

Here $\mathbb{H}^1$ in (6.0.9a) is a fourth-order symmetric tensor

$$(\mathbb{H}^1)_{ij} = \left(\frac{\partial^2 W}{\partial \mathbf{v}^2} \right)_{ij} = \frac{\partial^2 W}{\partial v_i \partial v_j},\tag{6.0.10}$$

where $i, j = 1, 2, 3, 4$, and $\mathbf{v} = (\lambda_2, \lambda_3, P, M)^T$.

Combining (6.0.8) - (6.0.10), we can recast the second variation in a more compact form

$$\delta^2 W^t(\mathbf{v}) = \delta \mathbf{v} \cdot \mathbb{H} \delta \mathbf{v},\tag{6.0.11}$$

where $\mathbb{H}$ is a fourth-order symmetric tensor

$$\mathbb{H} = \frac{\partial^2 W^t}{\partial \mathbf{v}^2} = \begin{pmatrix} \frac{\partial^2 W^t}{\partial \lambda_2^2} & \frac{\partial^2 W^t}{\partial \lambda_2 \partial \lambda_3} & \frac{\partial^2 W^t}{\partial \lambda_2 \partial P} & \frac{\partial^2 W^t}{\partial \lambda_2 \partial M} \\ & \frac{\partial^2 W^t}{\partial \lambda_3^2} & \frac{\partial^2 W^t}{\partial \lambda_3 \partial P} & \frac{\partial^2 W^t}{\partial \lambda_3 \partial M} \\ & & \frac{\partial^2 W^t}{\partial P^2} & \frac{\partial^2 W^t}{\partial P \partial M} \\ & \text{Sym} & & \frac{\partial^2 W^t}{\partial M^2} \end{pmatrix} \quad (6.0.12)$$

with the entries

$$\frac{\partial^2 W^t}{\partial \lambda_2^2} = \frac{\partial^2 W}{\partial \lambda_2^2} - \epsilon_0 (\lambda_3 \xi_{,X})^2 + \mu_0 (\lambda_3 \zeta_{,X}^{\text{self}})^2, \quad (6.0.13a)$$

$$\frac{\partial^2 W^t}{\partial \lambda_2 \partial \lambda_3} = \frac{\partial^2 W}{\partial \lambda_2 \partial \lambda_3} - 2\epsilon_0 \lambda_2 \lambda_3 (\xi_{,X})^2 + P \xi_{,X} + 2\mu_0 \lambda_2 \lambda_3 (\zeta_{,X}^{\text{self}})^2, \quad (6.0.13b)$$

$$\frac{\partial^2 W^t}{\partial \lambda_2 \partial P} = \frac{\partial^2 W}{\partial \lambda_2 \partial P} + \lambda_3 \xi_{,X}, \quad (6.0.13c)$$

$$\frac{\partial^2 W^t}{\partial \lambda_2 \partial M} = \frac{\partial^2 W}{\partial \lambda_2 \partial M} + 2\mu_0 \lambda_2 \lambda_3^2 \zeta_{,X}^{\text{self}} [\zeta_{,X}^{\text{self}}]_{,M}, \quad (6.0.13d)$$

$$\frac{\partial^2 W^t}{\partial \lambda_3^2} = \frac{\partial^2 W}{\partial \lambda_3^2} - \epsilon_0 (\lambda_2 \xi_{,X})^2 + \mu_0 (\lambda_2 \zeta_{,X}^{\text{self}})^2, \quad (6.0.13e)$$

$$\frac{\partial^2 W^t}{\partial \lambda_3 \partial P} = \frac{\partial^2 W}{\partial \lambda_3 \partial P} + \lambda_2 \xi_{,X}, \quad (6.0.13f)$$

$$\frac{\partial^2 W^t}{\partial \lambda_3 \partial M} = \frac{\partial^2 W}{\partial \lambda_3 \partial M} + 2\mu_0 \lambda_2^2 \lambda_3 \zeta_{,X}^{\text{self}} [\zeta_{,X}^{\text{self}}]_{,M}, \quad (6.0.13g)$$

$$\frac{\partial^2 W^t}{\partial P^2} = \frac{\partial^2 W}{\partial P^2}, \quad (6.0.13h)$$

$$\frac{\partial^2 W^t}{\partial P \partial M} = \frac{\partial^2 W}{\partial P \partial M}, \quad \text{and} \quad (6.0.13i)$$

$$\frac{\partial^2 W^t}{\partial M^2} = \frac{\partial^2 W}{\partial M^2} + \mu_0 (\lambda_2 \lambda_3)^2 [\zeta_{,X}^{\text{self}}]_{,M}^2. \quad (6.0.13j)$$

